Second-order response Bethe-Salpeter equation

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A Bethe-Salpeter equation for the three-particle correlation function is derived, representing a second-order response generalization of the usual Bethe-Salpeter equation. The equation can be solved formally, giving the three-particle correlation function in terms of the two-particle correlation function and many-body interaction kernels. The similarity to second-order response time-dependent density-functional theory can be used to improve the understanding of the higher-order exchange and correlation kernels of this theory. An exact expression for such a kernel is derived.

I. INTRODUCTION

The Bethe-Salpeter equation (BSE) describing two-particle bound states within quantum field theory [1,2] can be considered as one of the fundamental equations of modern physics. It has a vast spectrum of applications, ranging from nuclear physics [3], atomic physics [4], to solid-state physics [5] and physical chemistry [6]. Apart from being a fundamental equation, its success comes from the fact that in its formulation in terms of the two-particle correlation functions [1,7,8], the non-interacting correlation function can be readily written down in many cases and the interaction enters via a kernel that is the variation of the self-energy and thus is suitable for intuitive physical approximations within many-body theory. As pointed out by Baym and Kadanoff [7], however, the two-particle correlation function describes only phenomena in a linear response regime, i.e., to first order in an external perturbation.

A comprehensive understanding of second-order perturbation processes under many-body conditions is, however, important in many fields, such as atomic physics [9], nonlinear optics [10,11], solid-state physics [12], to statistical physics [13]. Second-order responses are intimately linked to three-particle correlations, which in analogy to the linear case, contain all second-order properties, and thus represent the basic quantity describing these processes.

A generalization of the BSE to the regime of nonlinear response phenomena has, to the best of my knowledge, not yet been undertaken. Such a generalization amounts to formulate a connection between the noninteracting and interacting three-particle correlation functions. As such, a second-order BSE describes the physics of second-order response phenomena with the same level of generality as the (linear) Bethe-Salpeter and should therefore be the natural starting point for systematic approximations for the many-body physics of second order phenomena. Formally the BSE is a Dyson equation that includes the interaction to infinite order and thus gives an interacting linear response quantity. Likewise, a second-order BSE should contain the three-particle interaction to infinite order and give a second-order interacting response quantity. (It is important to note that in this context "second order"

refers to the external perturbation and not to an expansion in the interaction, as carried out by diagrammatic expansion, which is here always contained up to infinite order.)

In solid-state physics and physical chemistry linear response functions are routinely calculated within time-dependent density-functional theory (TDDFT) [14,15]. In this framework there does exist a relation that can be considered the second-order generalization of the Dyson equation. It relates the interacting second-order density response to its noninter-acting one via the exchange and correlation kernels f_{xc} and g_{xc}, which are the first- and second-order variations of the exchange and correlation potential of TDDFT [16,17]. The exact form of the linear interaction kernel f_{xc} is, however, unknown and finding good approximations to them remains one of the major challenges of the field [18–20]. The second-order kernel g_{xc}, being the variation of the linear one, is even less known and it has been virtually never used in practical calculations so far. The second-order TDDFT Dyson-like equation where this kernel features has been published some time ago [16] but has also never been applied until recently [17,21,22], arguably because of the lack of knowledge about g_{xc}.

On the other hand, some progress has been made regarding the linear interaction kernel f_{xc} by comparing the TDDFT Dyson equation with the BSE. Given that they have the same structure and that the two-particle correlation function can be contracted to yield a response function, an exact expression for f_{xc} can be obtained, describing two-particle many-body processes [18,19,23–26]. It is the underlying structure of the Dyson equation that both have in common and makes this comparison possible. In the same way, there is a correspondence of the structure between the known second-order TDDFT Dyson-like equation [16,17] and a second-order generalization of the BSE. Such a correspondence can not only guide the derivation of such an equation but also can serve to yield an expression for g_{xc} in terms of many-body quantities.

The aim of this paper is thus twofold: First, to establish the second-order BSE, which is a general expression applicable to many different physical phenomena in second-order perturbation regimes, just as the BSE does for the linear regime. Second, to demonstrate how this equation can be used to improve the existing understanding of second-order response TDDFT, in the sense that it gives an exact expression for the thus far largely unknown second-order interaction kernel g_{xc}.

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The outline of this paper follows these two aims. In Sec. II, I show how the known derivation of the linear BSE in many-body perturbation theory can be readily generalized to second order. This amounts to a rather straightforward second functional derivative. I also show how, given that the solution of the linear BSE is known, the second-order BSE can be solved formally, without an additional inversion step, in Sec. II A. Having established the form and solution of the second-order BSE, I relate it to the second-order TDDFT Dyson-like equation in Sec. III, which has the same form and solution. The combination of both equations yield an exact equation for g_{xc}, including all second-order many-body interactions as shown in Sec. IV.

II. DERIVATION OF THE SECOND-ORDER BSE

The Bethe-Salpeter equation provides an approach to many-body excitations within the framework of many-body perturbation theory [27]. More precisely it is an equation for the correlation part L_2 of the two-particle Green's function that can be written as [here and in the following I use the notation $G_1 = G(\bullet, \bullet)$, $G_2 = G(\bullet, \bullet, \bullet, \bullet)$, etc., i.e., whether a quantity is second order, of two particles, of three particles, etc., is determined by the number of variables]

$$i L(1,2,3,4) = -G(1,3,2,4) + G(1,2)G(3,4), \qquad (1)$$

where the product of the one-particle Green's functions $G_1 G_1$ describes the independent propagation of the two particles. Thus, the two-particle correlation function describes those parts of two-particle processes that go beyond their independent propagation that is represented by $G_1 G_1$. In many-body perturbation theory, this quantity is also defined as the variation of the one-particle Green's function under the presence of a perturbing potential using Schwinger's functional derivative [27,28]

$$L(1,2,3,4) = -i \frac{\delta G(1,2)}{\delta V_{\text{per}}(3,4)}, \qquad (2)$$

while the single particle G is determined by the Dyson equation

$$G^{-1}(1,2) = G_H^{-1}(1,2) - V_{\text{per}}(1,2) - \Sigma(1,2), \qquad (3)$$

where Σ is the self-energy and G_H is the Hartree Green's function [27,29]. Combining the two equations and carrying out the functional derivative yields the BSE in the form [2]

$$L(1,2,3,4) = L_0(1,2,3,4) + \int d5 \, d6 \, d7 \, d8 \, L_0(1,2,5,6)$$
$$\times \tilde{\Xi}(5,6,7,8)L(7,8,3,4), \qquad (4)$$

where the many-body interaction kernel $\tilde{\Xi}$ is defined as

$$\tilde{\Xi}(5,6,7,8) = v(5,7)\delta(5,6)\delta(7,8) + i\frac{\delta \Sigma(5,6)}{\delta G(7,8)} \qquad (5)$$

and the noninteracting part of the correlation function is

$$L_0(1,2,3,4) = -iG(1,3)G(4,2). \qquad (6)$$

For second-order processes in response formulation, one is interested in the quadratic response, i.e., the response to two

perturbing fields. Thus, generalizing Eq. (2) to second order, one finds the corresponding three-particle correlation function

$$L(1,2,3,4,5,6) = -i\frac{\delta^2 G(1,2)}{\delta V_{\text{per}}(5,6)\delta V_{\text{per}}(3,4)} = \frac{\delta L(1,2,3,4)}{\delta V_{\text{per}}(5,6)}. \qquad (7)$$

This quantity can be interpreted as the correlation part of the three-particle Green's function, which is obtained from Eq. (1) by taking the functional derivative with respect to an additional nonlocal perturbing potential

$$iL(1,2,3,4,5,6) = -G(1,3,5,2,4,6) - G(1,3,2,4)G(5,6)$$
$$- G(1,5,2,6)G(3,4) - G(3,5,4,6)G(1,2)$$
$$+ 2G(1,2)G(3,4)G(5,6). \qquad (8)$$

There are not only the free propagations of three particles represented by the $G_1 G_1 G_1$ term but also the fully interacting propagation of pairs of particles with an independent third one represented by $G_2 G_1$. One can thus see from this equation that L_3 indeed represents the three-particle correlation part of G_3.

A second-order BSE is now readily obtained by carrying out an additional functional derivative of Eq. (4)—the steps involved are straightforward but cumbersome because of the amount of indices involved, so that the details are given in the Appendix A. The final result reads [cf. Eq. (A7) for indices]

$$L_3 = L_{03} + L_{03}\tilde{\Xi}_2 L_2 + L_{03}\tilde{\Xi}_2 L_2 + L_{03}\tilde{\Xi}_2 L_2 \tilde{\Xi}_2 L_2$$
$$+ L_{02}\Xi_3 L_2 L_2 + L_{02}\tilde{\Xi}_2 L_3, \qquad (9)$$

where the noninteracting part L_{03} is defined as

$$iL_0(123456) = G(1,5)G(6,3)G(4,2) + G(1,3)G(4,5)G(6,2) \qquad (10)$$

and the second-order many-body interaction kernel reads

$$\Xi(1,2,3,4,5,6) = i\frac{\delta^2 \Sigma(1,2)}{\delta G(5,6)\delta G(3,4)}. \qquad (11)$$

Equation (9) is an equation for the second-order quantity L_3 but it also contains the first-order correlation function and L_2 and interaction kernel Ξ_2, i.e., there are contributions that depend only on two-particle interaction. The structure of Eq. (9) can be seen as the second-order equivalent of the Dyson equation and it is the same encountered in second-order response TDDFT [16,17].

A. Formal solution

An important feature of the second-order Dyson-like equation is that if the solution of the corresponding first order is known, its solution can be formally written down. Again, this involves rather cumbersome indices, so here I will show only a shorthand version, while I give the full index dependence in Appendix A.

Equation (9) can be rearranged to

$$[1 - L_{02}\tilde{\Xi}_2]L_3 = L_{03}[1 + \tilde{\Xi}_2 L_{02}][1 + \tilde{\Xi}_2 L_{02}] + L_{02}\Xi_3 L_2 L_2 \qquad (12)$$

and the first-order BSE (4) can be written as

$$[1 - L_{02}\tilde{\Xi}_2] = L_{02}L_2^{-1} \qquad (13)$$

and

$$[1 + \tilde{\Xi}_2 L_{02}] = L_2 L_{02}^{-1}. \qquad (14)$$

Inserting these last two expressions in Eq. (12) and multiplying from the left with $L_{02}^{-1} L_2$ solves the second-order BSE:

$$L_3 = L_2 L_{02}^{-1} L_{03} L_{02}^{-1} L_2 L_{02}^{-1} L_2 + L_2 \Xi_3 L_2 L_2, \qquad (15)$$

or alternatively

$$L_3 = [1 + L_2 \tilde{\Xi}_2] L_{03} [1 + \tilde{\Xi}_2 L_2][1 + \tilde{\Xi}_2 L_2] + L_2 \Xi_3 L_2 L_2. \qquad (16)$$

While obtaining this solution appears formally rather trivial, it is an important point to notice that to evaluate this solution one does not need to invert the second-order quantities, which are in this case six-point quantities and thus very demanding to handle numerically. The three-particle correlation function thus just needs to be evaluated using the linear result, the noninteracting three-particle propagator L_{03}, and the three-particle interaction kernel Ξ_3. Still, such an evaluation is numerically challenging because first one needs to construct L_{03}, which is a six-point quantity as well, and second, as shown in Eq. (A12), it involves the integral (or sum) over 12 indices, which leads to very unfavorable scaling properties.

B. The second-order many-body kernel Ξ_3

The linear many-body interaction kernel is the variation of the self-energy with respect to a single-particle Green's function. In practical applications in solid-state physics of the BSE this self-energy is taken in Hedin's GW approximation [30,31], so that the kernel reads

$$\begin{aligned} \Xi(5,6,7,8) &= -\frac{\delta[G(5,6)W(5,6)]}{\delta G(7,8)} \\ &= -\delta(5,7)\delta(6,8)W(5,6) - G(5,6)\frac{\delta W(5,6)}{\delta G(7,8)}. \end{aligned} \qquad (17)$$

Additionally, one assumes that the functional derivative of the screening W with respect to the Green's function $\delta W/\delta G$, which describes the change of the screening due to the excitation, is small and can thus be neglected, as shown by Hanke and Sham [32]. This assumption is, however, an *ad hoc* approximation and mainly justified pragmatically. In this approximation the second-order kernel that is defined as

$$\Xi(1,2,3,4,5,6) = i\frac{\delta^2 \Sigma(1,2)}{\delta G(5,6)\delta G(3,4)} = \frac{\delta \Xi(1234)}{\delta G(5,6)} \qquad (18)$$

obviously vanishes as well. It does, however, not vanish *a priori* if one considers other approximations for the first-order kernel—cf. Ref. [7], for example.

Furthermore, even in GW, the assumption $\delta W/\delta G = 0$ could mean that one is missing important contributions and it might not be a good approximation when one is interested in second-order processes. Especially, since the second-order BSE is describing second-order processes, this term that is second order in W and is describing the change of the screening due to the excitation can be important.

In atomic physics and chemistry one usually only considers a Hartree-Fock self-energy of the form $\Sigma = iGv$, for which

the second-order interaction kernel vanishes as well. We will see below, however, that even if the kernel vanishes, there are still finite second-order processes beyond the independent propagation remaining.

III. RELATION TO TDDFT

The time-dependent density-functional theory in response formulation relates the interacting density responses χ to the noninteracting Kohn-Sham responses χ_0 via the exchange and correlation kernels f_{xc} [14],

$$\chi(1,2) = \chi_0(1,2) + \int d3\, d4 \chi_0(1,3) f_{vxc}(3,4)\chi(4,2), \qquad (19)$$

where $f_{vxc} = v + f_{xc}$. This is in the same way as the BSE relates correlation function L_2 to the noninteracting part via the many-body interaction kernel Ξ_2. The underlying common structure is the Dyson equation. A second-order response equivalent of the TDDFT Dyson equation is given in Refs. [16,17,21] and reads

$$\begin{aligned} \chi_2 &= \chi_{02} + \chi_{02} f_{vxc} \chi_1 + \chi_{02} f_{vxc} \chi_1 + \chi_{02} f_{vxc} \chi_1 f_{vxc} \chi_1 \\ &\quad + \chi_{01} g_{xc} \chi_1 \chi_1 + \chi_{01} f_{vxc} \chi_2 \end{aligned} \qquad (20)$$

and yields exactly the same structure as the second-order BSE, Eq. (9). This seems to be the general structure of how second-order quantities are related to their "noninteracting" counterparts via interaction kernels and as such can be regarded as second-order Dyson equations.

The second-order TDDFT Dyson equation can also be solved formally in the same way as the second-order BSE. One finds

$$\chi_2 = \chi_1 \chi_{01}^{-1} \chi_{02} \chi_{01}^{-1} \chi_1 \chi_{01}^{-1} \chi_1 + \chi_1 g_{xc} \chi_1 \chi_1 \qquad (21)$$

and equivalently

$$\chi_2 = [1 + \chi_1 f_{vxc}]\chi_{02}[1 + f_{vxc}\chi_1][1 + f_{vxc}\chi_1] + \chi_1 g_{xc}\chi_1\chi_1. \qquad (22)$$

These are again shorthand versions, the full indexed versions of which are given in Appendix B.

Having established the formal similarities between the second-order TDDFT and BSE, it is important to note their fundamental differences. As already pointed out above, the fundamental quantity of the BSE is the one-particle Green's function and its variation with respect to a perturbing potential yields the two-particle correlation function according to Schwinger's relation. The fundamental quantity of TDDFT is the density and its variation with respect to a perturbing potential yields the density response function. By definition, the Green's function is nonlocal in space and time, since it describes the propagation of a particle, while, also by definition, the density is a local property. Formally, this means that the BSE is an equation of four-point (or six-point) quantities and TDDFT of two-point (or three-point) quantities. The two are connected, however, by the fact that the contraction of the Green's function yields the density $iG(1,1') = \rho(1)$. In this sense all the properties of the density response χ_1 (or χ_2) are contained in the two-particle correlation L_2 (or three-particle correlation L_3) and can be accessed by

contraction, i.e.,

$$L(1,1',2,2') = -i\frac{\delta G(1,1')}{\delta V_{per}(2,2')} = \frac{\delta \rho(1)}{\delta V_{per}(2)} = \chi(1,2) \quad (23)$$

and

$$L(1,1',2,2',3,3') = -i\frac{\delta G(1,1')}{\delta V_{per}(2,2')\delta V_{per}(3,3')}$$
$$= \frac{\delta^2 \rho(1)}{\delta V_{per}(2)\delta V_{per}(3)} = \chi(1,2,3). \quad (24)$$

This does not, however, mean that the TDDFT Dyson equation is a trivial contraction of the BSE, which is not the case [5,18]. Indeed, in the linear case one first has to solve the four-point BSE to obtain L_2 that then can be contracted to give the linear density response.

This contraction property also holds for the noninteracting quantities, which means that the independent (quasiparticle-corrected) density response can be written as

$$\chi_0(1,2,3) = L_0(1,1',2,2',3,3')$$
$$= -iG(1,2)G(2,3)G(3,1) - iG(1,3)G(3,2)G(2,1). \quad (25)$$

This is also the second-order generalization of the linear random phase approximation (RPA) polarizability $P_1 = -iG_1G_1$. We note that for second order two separate terms are necessary to assure symmetry in the indices of the perturbation, i.e., $2 \leftrightarrow 3$.

Another way of interpreting Eq. (25) is that it represents a particle-particle-hole and a particle-hole-hole process. Note that this depends on contraction chosen here, and other contractions imply different time ordering and thus can yield, e.g., hole-hole-hole processes, etc.

In this paper we only discuss density responses, but L_2 and L_3 contain much more information than that. They can, for example, also be used to give the current response, as shown by Gatti [33] for the linear case.

IV. A g_{xc} FROM MANY-BODY PERTURBATION THEORY

The similarity between the BSE and the fact that the contraction of L_2 yields the linear density response has been used to derive an exact expression for the two-particle correlation part of the TDDFT kernel f_{xc} [18]. This derivation uses the fact that the kernel can be written as consisting of two parts, $f_{xc} = f_{xc}^{(1)} + f_{xc}^{(2)}$, where $f_{xc}^{(1)}$ accounts for the quasiparticle corrections and $f_{xc}^{(2)}$ for other many-body effects [25]. Following the derivation of Gatti *et al.* [20] for the first order, I will here sketch how the second-order BSE can be used to derive a similar expression for g_{xc}.

Since the Coulomb interaction v is known and does not contribute directly to f_{xc} and g_{xc}, it is convenient to compare only the irreducible quantities, here denoted P and $\tilde{L}$, in TDDFT and BSE, respectively. The relations connecting the irreducible polarizabilities P with the reducible response functions χ are

$$P_1 = \chi_1 + \chi_1 v P_1, \quad (26)$$
$$P_2 = [1 - P_1 v]\chi_2[1 - vP_1][1 - vP_1] \quad (27)$$
$$= P_1\chi_1^{-1}\chi_2\chi_1^{-1}P_1\chi_1^{-1}P_1, \quad (28)$$

and similar for L and $\tilde{L}$. The solutions of the second-order Dyson equations for these quantities read

$$P_2 = P_1P_{01}^{-1}P_{02}P_{01}^{-1}P_1P_{01}^{-1}P_1 + P_1g_{xc}P_1P_1, \quad (29)$$
$$L_3 = \tilde{L}_2\tilde{L}_{02}^{-1}L_{03}\tilde{L}_{02}^{-1}\tilde{L}_2\tilde{L}_{02}^{-1}\tilde{L}_2 + \tilde{L}_2\Xi_3\tilde{L}_2\tilde{L}_2. \quad (30)$$

Here I only give the shorthand notation, but it is understood that $\tilde{L}_2$ and $\tilde{L}_3$ are four- and six-point quantities, while P_1 and P_2 are two- and three-point quantities.

The two equations can be combined by exploiting the similarity of the two independent particle responses L_{03} and P_{02}. As shown in Sec. III, the three-point contraction of L_{03} equals the independent density response and thus also P_{02}. Since the BSE cannot be contracted to yield the TDDFT Dyson equation, one has to generalize instead Eq. (29) to six points, thus making all P and P_0 trivially contractible four- (^4P) and six-point (^6P) quantities—cf. Appendix C.

Now, using $L_{03} = {}^6P_{02}$ and $L_{02} = {}^4P_{01}$, the two equations can be combined and solved for the kernels

$$\Xi_3 - {}^6g_{xc} = \tilde{L}_2^{-1}\tilde{L}_3\tilde{L}_2^{-1}\tilde{L}_2^{-1} - {}^4P_1^{-1}{}^6P_2\,{}^4P_1^{-1}{}^4P_1^{-1}. \quad (31)$$

At this point we already note that even in the cases where $\Xi_3 = 0$, cf. Sec. II B, the second-order TDDFT kernel g_{xc} is still finite. Indeed, it only vanishes if additionally ${}^6P_2 = \tilde{L}_3$ and ${}^4P_1 = \tilde{L}_2$, which is generally false and can only be achieved in oversimplified models. In particular, this means that in the GW approximation with the additional assumption of $\delta W/\delta G = 0$, where $\Xi_3 = 0$, the second-order TDDFT kernel g_{xc} generally does not vanish. That means that g_{xc} has not only to account for interactions that are purely of second order in the sense of the Bethe-Salpeter interactions kernels, i.e., three-particle interaction, but also has to account for some nontrivial coupling of linear quantities.

This is, for example, the case in the Hartree-Fock approximation, because it does not consider correlation but only accounts for exchange which is by definition a two-particle process. Still, in this approximation g_{xc} is nonzero, which clearly shows that the folding of many-body processes into the effective kernels f_{xc} and g_{xc} reduces the intuitivity of TDDFT for higher-order processes. It also means that in TDDFT calculations that let $g_{xc} = 0$ not only three-particle correlations are neglected, but also one misses exchange effects.

To get an exact expression for g_{xc}, we can now use the fact that P_2 is the three-point contraction of $\tilde{L}_3$ and thus let ${}^6P_2|_{6\to3} = P_2 = \tilde{L}_3|_{6\to3}$. By solving Eq. (31) for L_3, contracting the free indices and making this substitution, we obtain

$$P_2 = \left(\tilde{L}_2\,{}^4P_1^{-1}\,{}^6P_2\,{}^4P_1^{-1}\tilde{L}_2\,{}^4P_1^{-1}\tilde{L}_2\right)\Big|_{6\to3}$$
$$+ \left(\tilde{L}_2(\Xi_3 - {}^6g_{xc})\tilde{L}_2\tilde{L}_2\right)\Big|_{6\to3}, \quad (32)$$

where $6 \to 3$ indicates the pairwise contraction of the six free indices to three. This constitutes a generalized Sham-Schlüter equation [20,29] for the kernels that now can be solved for g_{xc}. To keep track of the contracted quantities, it is necessary to explicitly account for the indices while proceeding—cf. Appendix C. Therefore, the resulting expression (C9) lacks readability, so that here I give again only a shorthand, indicating left-hand or right-hand-sided contractions of four-point quantities as ${}_3|\bullet$ and $\bullet|_3$, respectively. Thus, Eq. (32)

solved for g_{xc} reads

$$g_{xc} = P_1^{-1} \left[\,_3 \big| \tilde{L}_2 \,^4 P_1^{-1} \big|_3 P_2 \,_3 \big| \,^4 P_1^{-1} \tilde{L}_2 \big|_3 \,_3 \big| \,^4 P_1^{-1} \tilde{L}_2 \big|_3 - P_2 \right]$$
$$\times P_1^{-1} P_1^{-1} + P_1^{-1} \,_3 \big| \tilde{L}_2 \Xi_3 \tilde{L}_2 \big|_3 \tilde{L}_2 \big|_3 P_1^{-1} P_1^{-1}. \quad (33)$$

In comparison, the corresponding expression of the linear f_{xc} derived in this framework (cf. Refs. [20] and [29]) reads in this notation

$$f_{xc} = P_1^{-1} \,_3 \big| \,^4 P_1 \Xi_2 L_2 \big|_3 P_1^{-1}. \quad (34)$$

To illustrate the notation in Eq. (33), we have, for example, quantities as

$$_3 \big| \,^4 P_1^{-1} \tilde{L}_2 \big|_3 = \,^4 P^{-1}(1,1,3,4) \tilde{L}(4,3,2,2), \quad (35)$$

from which we can see again that only if $^4 P_1 = \tilde{L}_2$, one can follow $g_{xc} = \Xi_3|_{6 \to 3}$. Instead, Eq. (33) gives the exact expression for a TDDFT kernel that reproduces a P_2 such that $P(1,2,3) = \tilde{L}(1,1,2,2,3,3,)$, i.e., a second-order irreducible polarizability that accounts for all three-particle many-body interactions. The advantage is that one does not have to solve the six-point second-order BSE but can keep the three-point formalism of second-order response TDDFT.

The downside is that, apart from having to perform a linear BSE calculation first to obtain $\tilde{L}_2$, the actual knowledge of the kernel g_{xc} as in Eq. (33) implies knowledge of P_2 and is therefore not possible. For calculation purposes one has to make an approximation on this equation, particularly on this instance of P_2. The most straightforward would be to let $P_2 \to P_{02}$.

One can also use this result to deduce some general properties a g_{xc} should have. For optical applications, for example, the behavior of this kernel in the optical limit $\mathbf{q} \to 0$ could be of interest. This limit can be readily obtained by realizing that in the exact expression of g_{xc}, Eq. (C9), the only free indices are those of P_1^{-1}. The $\mathbf{q}$ dependence of these quantities is known to be $\sim 1/q^2$, and since one of the two indices of these three P_1^{-1} is integrated, the overall dependence of g_{xc} in the optical limit is

$$\lim_{\mathbf{q} \to 0} g_{xc}(\mathbf{q}, \mathbf{q}, \mathbf{q}) \sim \frac{1}{q^3}. \quad (36)$$

The factor of proportionality can be seen as a material-dependent coefficient and Eq. (33) as the prescription on how to calculate it. In this sense the second-order BSE can improve the understanding and design of TDDFT kernels.

V. CONCLUSION

In this paper I have shown how a Bethe-Salpeter equation for the nonlinear response regime can be derived and that

it can be solved formally for the three-particle correlation function, provided the two-particle correlation is known. This result is general in the sense that is does not make any assumption on the underlying physical system, i.e., it should be applicable to the range of physical processes where many-body effects determine the nonlinear response of a system. Even more general, it provides in principle a prescription on how the three-particle correlation function can be obtained from the corresponding two-particle correlation functions. Physical approximations enter this solution mainly via the many-body self-energy, for which a vast amount of approximations and theories exist that could be directly applied to this equation.

Formally, this equation has the same structure as the second-order response TDDFT Dyson equation. This similarity points to some underlying general structure for second-order quantities in Dyson-like equations. It can also be systematically exploited to improve the current understanding of TDDFT as far the second-order exchange and correlation kernel is concerned. It turns out that there is no direct correspondence between this kernel and the three-particle interaction kernel of the BSE, but the comparison of TDDFT and BSE makes it possible to disentangle the many-body effects that are contained in g_{xc}, as shown, for example, in the Hartree-Fock approximation, where g_{xc} contains two-particle exchange effects. Furthermore, one can derive an exact expression for a g_{xc} that reproduces a polarizability with the same many-body features as the three-particle correlation function. This expression provides the possibility to deduce some general properties of g_{xc}, such as its $\mathbf{q}$ dependence in the optical limit, so that it can be a starting point for systematic approximations for this kernel.

The extensive discussion of the relation between the second-order BSE and the TDDFT formalism is, however, only one of many possible fields where this equation can provide further insight into higher-order physics.

Note added in proof. Recently, I have become aware of a work [34] that deals with a similar formulation.

APPENDIX A: DETAILS OF DERIVATION OF THE SECOND-ORDER BSE

To obtain a Bethe-Salpeter-like equation for the three-particle correlation function L_3, we simply have to derive the first-order BSE, Eq. (4):

$$\frac{\delta L(1,2,3,4)}{\delta V_{\text{per}}(5,6)} = \frac{\delta}{\delta V_{\text{per}}(5,6)} [-i G(1,3) G(4,2)] + \frac{\delta}{\delta V_{\text{per}}(5,6)} \left\{ \int d7 \, d8 \, d9 \, d10 (-i) G(1,7) G(8,2) \right.$$
$$\times \left[v(7,9) \delta(7,8) \delta(9,10) + i \frac{\delta \Sigma(7,8)}{\delta G(9,10)} \right] (-i) \frac{\delta G(9,10)}{\delta V_{\text{per}}(3,4)} \right\} = (-i) \frac{\delta G(1,3)}{\delta V_{\text{per}}(5,6)} G(4,2) + (-i) G(1,3) \frac{\delta G(4,2)}{\delta V_{\text{per}}(5,6)}$$
$$+ \int d7 \, d8 \, d9 \, d10 (-i) \frac{\delta G(1,7)}{\delta V_{\text{per}}(5,6)} G(8,2) \left[v(7,9) \delta(7,8) \delta(9,10) + i \frac{\delta \Sigma(7,8)}{\delta G(9,10)} \right] (-i) \frac{\delta G(9,10)}{\delta V_{\text{per}}(3,4)}$$

$$+ \int d7\,d8\,d9\,d10\,(-i)G(1,7)\frac{\delta G(8,2)}{\delta V_{\text{per}}(5,6)}\left[v(7,9)\delta(7,8)\delta(9,10)+i\frac{\delta\Sigma(7,8)}{\delta G(9,10)}\right](-i)\frac{\delta G(9,10)}{\delta V_{\text{per}}(3,4)}$$

$$+ \int d7\,d8\,d9\,d10\,(-i)G(1,7)G(8,2)i\frac{\delta}{\delta V_{\text{per}}(5,6)}\left[\frac{\delta\Sigma(7,8)}{\delta G(9,10)}\right](-i)\frac{\delta G(9,10)}{\delta V_{\text{per}}(3,4)}$$

$$+ \int d7\,d8\,d9\,d10\,(-i)G(1,7)G(8,2)\left[v(7,9)\delta(7,8)\delta(9,10)+i\frac{\delta\Sigma(7,8)}{\delta G(9,10)}\right](-i)\frac{\delta^2 G(9,10)}{\delta V_{\text{per}}(5,6)\delta V_{\text{per}}(3,4)}. \tag{A1}$$

We note the repeated occurrence of first-order quantities known from first-order BSE. The only new term is the second derivative of the self-energy in the second last line. In this term we use the chain rule and get

$$\frac{\delta}{\delta V_{\text{per}}(5,6)}\left[\frac{\delta\Sigma(7,8)}{\delta G(9,10)}\right]=\int d11\,d12\frac{\delta}{\delta G(11,12)}\left[\frac{\delta\Sigma(7,8)}{\delta G(9,10)}\right]\frac{\delta G(11,12)}{\delta V_{\text{per}}(5,6)}. \tag{A2}$$

Using the definitions of the two-particle and three-particle many-body kernels, Eqs. (5) and (11), and inserting the known first-order quantities we can write

$$\frac{\delta L(1,2,3,4)}{\delta V_{\text{per}}(5,6)}=L(1,3,5,6)G(4,2)+G(1,3)L(4,2,5,6)+\int d7\,d8\,d9\,d10\,L(1,7,5,6)G(8,2)\tilde\Xi(7,8,9,10)L(9,10,3,4)$$

$$+\int d7\,d8\,d9\,d10\,G(1,7)L(8,2,5,6)\tilde\Xi(7,8,9,10)L(9,10,3,4)+\int d7\dots d12\,L_0(1,2,7,8)\Xi(7,8,9,10,11,12)$$

$$\times L(11,12,5,6)L(9,10,3,4)+\int d7\,d8\,d9\,d10\,L_0(1,2,7,8)\tilde\Xi(7,8,9,10)\frac{\delta L(9,10,3,4)}{\delta V_{\text{per}}(5,6)}. \tag{A3}$$

This is in principle already a second-order BSE. We note that at this point we do not need any six-point quantities other than the kernel.

To make the connection to the TDDFT Dyson equation and to avoid explicit reference to the one-particle Green's function G, we define

$$iL_0'(1,2,3,4,5,6)=G(1,3)G(4,2)G(5,6) \tag{A4}$$

and insert the full first-order expressions for the L_2 [Eq. (4)] in the above equation. Recalling the definition for $L_3=\delta L_2/\delta V$ we have (to keep the equation readable I drop the separating commas between variables, relying on the readers' goodwill to distinguish)

$$L(123456)=L_0'(135642)+L_0'(425613)+\int d7\,d8\,d9\,d10\,L_0'(137842)\tilde\Xi(78910)L(9\,10\,56)$$

$$+\int d7\,d8\,d9\,d10\,L_0'(427813)\tilde\Xi(78910)L(9\,10\,56)+\int d7\,d8\,d9\,d10\,L_0'(175682)\tilde\Xi(789\,10)L(9\,10\,34)$$

$$+\int d7\,d8\,d9\,d10\,L_0'(825617)\tilde\Xi(789\,10)L(9\,10\,34)+\int d7\,d8\,d9\,d10\,d11\,d12\,d13\,d14\,L_0'(17\,11\,12\,82)$$

$$\times\tilde\Xi(11\,12\,13\,14)L(13\,14\,56)\tilde\Xi(789\,10)L(9\,10\,34)+\int d7\,d8\,d9\,d10\,d11\,d12\,d13\,d14\,L_0'(82\,11\,12\,17)$$

$$\times\tilde\Xi(11\,12\,13\,14)L(13\,14\,56)\tilde\Xi(789\,10)L(9\,10\,34)+\int d7\,d8\,d9\,d10\,d11\,d12\,L_0(1278)\Xi(789\,10\,11\,12)$$

$$\times L(11\,12\,56)L(9\,10\,34)+\int d7\,d8\,d9\,d10\,L_0(1278)\tilde\Xi(789\,10)L(9\,10\,3456). \tag{A5}$$

We note that the eight first terms are in fact pairs of terms with the same structure. This is due to the symmetry in the perturbing fields, i.e., itdoes not make a physical difference if the $V_{\text{per}}(5,6)$ field is applied before the $V_{\text{per}}(3,4)$ field or vice versa. We can see that by exchanging the indices $3\leftrightarrow5$ and $4\leftrightarrow6$ in the equation. We therefore define an L_0 such that it accounts for these two possibilities:

$$iL_0(123456)=iL_0'(135642)+iL_0'(425613)=G(1,5)G(6,3)G(4,2)+G(1,3)G(4,5)G(6,2) \tag{A6}$$

with this the second order BSE reads

$$L(123456)=L_0(123456)+\int d7\,d8\,d9\,d10\,L_0(123478)\tilde\Xi(78910)L(9\,10\,56)+\int d7\,d8\,d9\,d10\,L_0(127856)\tilde\Xi(789\,10)L(9\,10\,34)$$

$$+\int d7\,d8\,d9\,d10\,d11\,d12\,d13\,d14\,L_0(1278\,11\,12)\tilde\Xi(11\,12\,13\,14)L(13\,14\,56)\tilde\Xi(789\,10)L(9\,10\,34)$$

$$+\int d7\,d8\,d9\,d10\,d11\,d12\,L_0(1278)\Xi(789\,10\,11\,12)L(11\,12\,56)L(9\,10\,34)$$

$$+\int d7\,d8\,d9\,d10\,L_0(1278)\tilde\Xi(789\,10)L(9\,10\,3456). \tag{A7}$$

To formally solve the second-order BSE we rearrange it to

$$\int d7\,d8\,d9\,d10\left[\delta(1,7)\delta(7,9)\delta(2,8)\delta(8,10) - L_0(1278)\tilde{\Xi}(78910)\right]L(9\,10\,3456)$$

$$= \int d7\ldots d14 L_0(1278\,11\,12)\left[\delta(7,3)\delta(8,4) + \tilde{\Xi}(78910)L(9\,10\,34)\right]\left[\delta(5,11)\delta(6,12) + \tilde{\Xi}(11\,12\,13\,14)L(13\,14\,56)\right]$$

$$+ \int d7\,d8\,d9\,d10\,d11\,d12 L_0(1278)\Xi(789\,10\,11\,12)L(11\,12\,56)L(9\,10\,34). \tag{A8}$$

Now we can use the linear BSE to write for the factor on the left-hand side,

$$\int d7\,d8\left[\delta(1,9)\delta(2,10) - L_0(1278)\tilde{\Xi}(78910)\right] = \int d7\,d8 L_0(1278)L^{-1}(87\,10\,9) \tag{A9}$$

as well as to rewrite the two linear factors on the right-hand side according to

$$\int d5\,d6\left[\delta(3,1)\delta(4,2) + \tilde{\Xi}(1256)L(5634)\right] = \int d5\,d6 L_0^{-1}(2165)L(5634), \tag{A10}$$

so that the second-order BSE can be written as

$$\int d7\,d8\,d9\,d10 L_0(1278)L^{-1}(87\,10\,9)L(9\,10\,3456)$$

$$= \int d7\ldots d14 L_0(1278\,11\,12)L_0^{-1}(87\,10\,9)L(9\,10\,34)L_0^{-1}(12\,11\,14\,13)L(13\,14\,56)$$

$$+ \int d7\,d8\,d9\,d10\,d11\,d12 L_0(1278)\Xi(789\,10\,11\,12)L(11\,12\,56)L(9\,10\,34). \tag{A11}$$

Now multiplying from the left with $\int d12\,d15\,d16 L(17\,18\,15\,16)L_0^{-1}(16\,15\,21)$ and renaming the indices $17 \leftrightarrow 1$ and $18 \leftrightarrow 2$ we have the final result:

$$L(123456) = \int d7\ldots d18 L(12\,15\,16)L_0^{-1}(16\,15\,18\,17)L_0(17\,18\,78\,11\,12)L_0^{-1}(8,7\,10\,9)L(9\,10\,34)L_0^{-1}(12\,11\,14\,13)L(13\,14\,56)$$

$$+ \int d7\,d8\,d9\,d10\,d11\,d12\,d15\,d16 L(12\,15\,16)\Xi(15\,16\,9\,10\,11\,12)L(11\,12\,56)L(9\,10\,34). \tag{A12}$$

APPENDIX B: TDDFT EQUATIONS

The full index-dependent versions of Eqs. (20)–(22) read

$$\chi(1,2,3) = \chi_0(1,2,3) + \int d4\,d5 \chi_0(1,4,3)f_{vxc}(4,5)\chi(5,2) + \int d4\,d5\chi_0(1,2,4)f_{vxc}(4,5)\chi(5,3)$$

$$+ \int d4\,d5\,d6\,d7\chi_0(1,5,4)f_{vxc}(5,6)\chi(6,2)f_{vxc}(4,7)\chi(7,3)$$

$$+ \int d4\,d5\,d6\chi_0(1,4)g_{xc}(4,5,6)\chi(6,3)\chi(5,2) + \int d4d5\chi_0(1,4)f_{vxc}(4,5)\chi(5,2,3), \tag{B1}$$

$$\chi(1,2,3) = \int d4\ldots d9\chi(1,8)\chi_0^{-1}(8,9)\chi_0(9,5,4)\chi_0^{-1}(5,6)\chi(6,2)\chi_0^{-1}(4,7)\chi(7,3) + \int d4\,d5\,d6\,d7\chi(1,7)g_{xc}(7,5,6)\chi(6,2)\chi(5,3), \tag{B2}$$

$$\chi(1,2,3) = \int d4\ldots d9\left[\delta(1,9) + \chi(1,8)f_{vxc}(8,9)\right]\chi_0(9,5,4)\left[\delta(2,5) + f_{vxc}(5,6)\chi(6,2)\right]\left[\delta(3,4) + f_{vxc}(4,7)\chi(7,3)\right]$$

$$+ \int d4\,d5\,d6\,d7\chi(1,7)g_{xc}(7,5,6)\chi(6,2)\chi(5,3). \tag{B3}$$

APPENDIX C: DETAILS OF DERIVATION OF g_{xc}

In this appendix I will give some details concerning the derivation of the second-order many-body exchange and correlation kernel g_{xc} as described in Sec. IV. To combine the second-order BSE and the TDDFT Dyson-like equation we have to represent

the TDDFT equation in terms of four- and six-point quantities, written as ^4P and ^6P. The Dyson-like equation for the irreducible polarizability reads

$$^6P(1,2,3,4,5,6) = \int d7\ldots d19\, {}^4P(1,2,7,8)\, {}^4P_0^{-1}(8,7,10,9)\, {}^6P_0(9,10,11,12,13,14)\, {}^4P_0^{-1}(12,11,16,17)\, {}^4P(16,17,3,4)$$
$$\times\, {}^4P_0^{-1}(14,13,19,18)\, {}^4P(18,19,5,6) + \int d7\ldots d12\, {}^4P(1,2,7,8)\, {}^6g_{xc}(7,8,9,10,11,12)$$
$$\times\, {}^4P(9,10,3,4)\, {}^4P(11,12,5,6), \tag{C1}$$

where

$$^6g_{xc}(7,8,9,10,11,12) = \delta(7,8)\delta(9,10)\delta(11,12)g_{xc}(7,9,11) \tag{C2}$$

and the linear quantities obey the four-point Dyson equation, as

$$^4P(1,2,3,4) = \int d5\, d6\, d7\, d8\, {}^4P_0(1,2,5,6)\left[\delta(3,5)\delta(4,6) + \delta(5,6)\delta(7,8)f(5,7)P(7,8,3,4)\right] \tag{C3}$$

so that

$$\int d12\, {}^4P_0^{-1}(10,9,2,1)\, {}^4P(1,2,3,4) = \int d7\, d8\left[\delta(3,9)\delta(4,10) + \delta(9,10)\delta(7,8)f(9,7)P(7,8,3,4)\right], \tag{C4}$$

and when one takes the contraction $P(1,1,2,2,3,3)$ of Eq. (C1) it collapses to

$$P(1,2,3) = \int d4\, d5\, d6\, d7\, d8\, d9\, P(1,4)P_0^{-1}(4,5)P_0(5,6,7)P_0^{-1}(6,8)P(8,2)P_0^{-1}(7,9)P(9,2)$$
$$+ \int d4\, d5\, P(1,4)g_{xc}(4,5,6)P(5,2)P(6,3). \tag{C5}$$

With these definitions the "Sham-Schlüter" equation (32) reads

$$\Xi(123456) - {}^6g_{xc}(123456) = \int d7\ldots d12\, \tilde{L}^{-1}(1278)\tilde{L}(789\,10\,11\,12)\tilde{L}^{-1}(10\,9\,34)\tilde{L}^{-1}(12\,11\,56)$$
$$- \int d7\ldots d12\, {}^4P^{-1}(1278)\, {}^6P(789\,10\,11\,12)\, {}^4P^{-1}(10\,9\,34)\, {}^4P^{-1}(12\,11\,56), \tag{C6}$$

and can be solved for $\tilde{L}_3$ to yield

$$\tilde{L}(13\,14\,15\,16\,17\,18) = \int d1\ldots d12\, \tilde{L}(13\,14\,21)\, {}^4P^{-1}(1278)\, {}^6P(789\,10\,11\,12)\, {}^4P^{-1}(10\,9\,34)\tilde{L}(43\,15\,16)\, {}^4P^{-1}(12\,11\,56)$$
$$\times\, \tilde{L}(65\,17\,18) + \int d1\ldots d6\, \tilde{L}(13\,14\,21)\left[\Xi(123456) - {}^6g_{xc}(123456)\right]\tilde{L}(43\,15\,16)\tilde{L}(65\,17\,18). \tag{C7}$$

Now, to use the property $\tilde{L}(13\,13\,15\,15\,17\,17) = P(13,15,17)$ we carry out this contraction, as well as collapsing all redundant indices and obtain

$$P(13,15,17) = \int d1\ldots d12\, \tilde{L}(13\,13\,21)\, {}^4P^{-1}(1277)P(79\,11)\, {}^4P^{-1}(9\,9\,34)\tilde{L}(43\,15\,15)\, {}^4P^{-1}(11\,11\,56)\tilde{L}(65\,17\,17)$$
$$+ \int d1\ldots d6\, \tilde{L}(13\,13\,21)\Xi(123456)\tilde{L}(43\,15\,15)\tilde{L}(65\,17\,17)$$
$$- \int d1\, d3\, d5\, \tilde{L}(13\,13\,11)g_{xc}(135)\tilde{L}(33\,15\,15)\tilde{L}(55\,17\,17). \tag{C8}$$

The diagonal $\tilde{L}_2$ in the last term are in fact two-point P_1 quantities, so that we obtain, solving this equation for g_{xc},

$$g_{xc}(1,2,3) = \int d4\ldots d15\, P^{-1}(1,4)[\tilde{L}(4456)\, {}^4P^{-1}(6577)P_2(7,8,9)\, {}^4P^{-1}(88\,11\,10)\tilde{L}(10\,11\,12\,12)$$
$$\times\, {}^4P^{-1}(99\,14\,13)\tilde{L}(13\,14\,15\,15) - P_2(4,12,15)]P^{-1}(12,2)P^{-1}(15,3)$$
$$+ \int d4\ldots d12\, P^{-1}(1,4)\tilde{L}(4456)\Xi(6587\,10\,9)\tilde{L}(78\,11\,11)\tilde{L}(9\,10\,12\,12)P^{-1}(12,2)P^{-1}(12,3). \tag{C9}$$

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