The density–density response function in time-dependent density functional theory: Mathematical foundations and pole shifting

Thiago Carvalho Corso, Mi-Song Dupuy, and Gero Friesecke

Abstract. We establish existence and uniqueness of the solution to the Dyson equation for the density–density response function in time-dependent density functional theory (TDDFT) in the random phase approximation (RPA). We show that the poles of the RPA density–density response function are forward shifted with respect to those of the noninteracting response function, thereby explaining mathematically the well-known empirical fact that the noninteracting poles (given by the spectral gaps of the time-independent Kohn–Sham equations) underestimate the true transition frequencies. Moreover, we show that the RPA poles are solutions to an eigenvalue problem, justifying the approach commonly used in the physics community to compute these poles.

1. Introduction

While ground state properties of molecules are very successfully captured by time-independent Kohn–Sham density functional theory (KS-DFT), excitation energies provide a much greater challenge. In particular, the excitation energies of the time-independent Kohn–Sham equations do not accurately capture the true excitation energies, and *have no theoretically supported meaning*.

Instead, time-dependent density functional theory (TDDFT) in the linear response regime has been found to capture a molecule's excitation spectrum much more accurately (see e.g. [24]). The underlying Dyson equation for the density–density response function of TDDFT has been derived as a meaningful approximation for this task (see e.g. [16,22]), and the overall approach has a huge physics literature. Our goal in this paper is to put TDDFT in the linear response regime and its connection with excitation spectra on a firm mathematical footing. We

    (1) establish existence and uniqueness of a solution to the Dyson equation for the density–density response function, in the basic case of the random phase approximation (RPA)

(2) mathematically clarify the relationship between the density–density response function and excitation spectra, by proving that the "exact" response function (coming from the evolution of the one-body density under full many-body quantum dynamics) has poles precisely at the excitation frequencies of the many-body Hamiltonian;

(3) show that the excitation frequencies obtained from the RPA Dyson equation are always forward shifted with respect to those of the time-independent Kohn–Sham equations.

Here, (1) proceeds by naturally viewing the density–density response function (DDRF) at a given time t as a linear operator between one-body potentials and identifying a suitable class of potentials on which the Dyson equation is well posed.

The result in (2) is considered "well known" in the physics literature. But the underlying Lehmann representation of the DDRF is not strictly speaking applicable to the molecular Hamiltonians to which one seeks to apply it in practice, as it tacitly assumes purely discrete spectrum and misses contributions from the continuous spectrum. Our advance is to provide a rigorous Lehmann representation of the DDRF which applies to molecular Hamiltonians and captures the contributions from the continuous spectrum.

Finally, (3) proceeds by characterizing the RPA poles as solutions to a certain eigenvalue problem, and carefully analyzing this eigenvalue problem.

Before stating our main results in more detail, let us introduce some background on linear response theory and on TDDFT.

1.1. Linear response theory

Linear response theory allows one to compute first-order corrections to quantities of interest of a molecule at equilibrium which is perturbed by an external potential. The exact wave function $\Psi(t)$ encoding the behavior of the electrons of the molecules is the solution to the time-dependent Schrödinger equation

$$\begin{cases} i\,\partial_t \Psi(t) = H(t)\Psi(t), & t > 0, \\ \Psi(0) = \Psi_0, \end{cases} \tag{1}$$

where

$$H(t) = H + \varepsilon f(t) V_{\mathcal{P}},$$

with $V_{\mathcal{P}}$ a time-independent bounded multiplicative potential (the *probe potential*) and f a bounded scalar function of time (the *time profile*). Here, Ψ_0 is the ground state of the rest Hamiltonian H, which for a molecule has the form

$$H = -\frac{1}{2}\Delta + \sum_{1 \le i < j \le N} w(r_i - r_j) + \sum_{i=1}^{N} v(r_i), \tag{2}$$

where $v, w \in L^2(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ and real valued.

For an observable $V_\mathcal{O}$ we are interested in the expectation value

$$\langle V_\mathcal{O} \rangle_t = \langle \Psi(t), V_\mathcal{O} \Psi(t) \rangle.$$

Since the perturbation is small, at first order in ε the variation of $\langle V_\mathcal{O} \rangle_t$ is

$$\langle V_\mathcal{O} \rangle_t = \langle V_\mathcal{O} \rangle_0 + \varepsilon (\mathcal{X}_{V_\mathcal{O} V_\mathcal{P}} \star f)(t) + \mathcal{O}(\varepsilon^2),$$

where $\mathcal{X}_{V_\mathcal{O} V_\mathcal{P}}$ is the function given by the Kubo formula (see Proposition 2.1)

$$\mathcal{X}_{V_\mathcal{O} V_\mathcal{P}}(\tau) = -i\theta(\tau) \langle V_\mathcal{O} \Psi_0, e^{-i(H-E_0)\tau} V_\mathcal{P} \Psi_0 \rangle + \text{c.c.}$$

This function has a Fourier transform, at least in the distributional sense,

$$\begin{aligned}
\widehat{\mathcal{X}}_{V_\mathcal{O} V_\mathcal{P}}(\omega) = \lim_{\eta \to 0^+} &\langle \Psi_0, V_\mathcal{O}(\omega + i\eta - (H - E_0))^{-1} V_\mathcal{P} \Psi_0 \rangle \\
&- \langle \Psi_0, V_\mathcal{P}(\omega + i\eta + (H - E_0))^{-1} V_\mathcal{O} \Psi_0 \rangle,
\end{aligned} \tag{3}$$

where $\eta \to 0^+$ means the one-sided limit as η converges to zero from above. This formula relates the singularities of the Fourier transform of $\widehat{\mathcal{X}}$ to the spectrum of H. More precisely, $\widehat{\mathcal{X}}$ has a pole if $|\omega|$ is an eigenvalue of $H - E_0$. When $|\omega|$ belongs to the essential spectrum of $H - E_0$, $\widehat{\mathcal{X}}$ is regular for a wide class of potentials v and w as a result of the limiting absorption principle. We refer to [1, 2, 6] for more information on this topic.

The location of the poles of the Fourier transform $\widehat{\mathcal{X}}$ provides access to the spectrum of H and in particular to its low-lying eigenvalues. At first sight, evaluating $\widehat{\mathcal{X}}$ from equation (3) is by no means simpler than diagonalizing the many-body operator H. However, a major simplification can be achieved, at least formally, by ("exact" and approximate) TDDFT, which are time-dependent versions of static Hohenberg–Kohn density functional theory [11, 13], respectively static Kohn–Sham density functional theory [12]. Provided the perturbing potential $V_\mathcal{P}$ and the observable $V_\mathcal{O}$ are *one-body potentials*

$$V_\mathcal{P} = \sum_{k=1}^{N} v_\mathcal{P}(r_k), \quad V_\mathcal{O} = \sum_{k=1}^{N} v_\mathcal{O}(r_k), \tag{4}$$

the expression for $\mathcal{X}$ becomes

$$\mathcal{X}_{V_\mathcal{O} V_\mathcal{P}}(\tau) = \langle v_\mathcal{O}, \chi(\tau) v_\mathcal{P} \rangle, \tag{5}$$

for some universal operator-valued function χ which is independent of $v_\mathcal{O}$ and $v_\mathcal{P}$ and only depends on the static Hamiltonian H in equation (2). This function, called the *density–density response function* of H, is rigorously constructed in Section 2.1. In the physics literature, χ is usually postulated to have an integral kernel, obtained formally by taking $v_\mathcal{O}$ and $v_\mathcal{P}$ to be delta functions located at r, respectively r',

$$\chi(r, r', \tau) = \langle \delta_r, \chi(\tau) \delta_{r'} \rangle,$$

and this kernel is known as the density–density response function. The operators $V_{\mathscr{P}}$ and $V_{\mathscr{O}}$ in (4) are then the density operators $\hat{\rho}_r = \sum_{i=1}^{N} \delta_r(r_i)$ and $\hat{\rho}_{r'} = \sum_{i=1}^{N} \delta_{r'}(r_i)$, explaining the name. Mathematically, there is no need for – or advantage from – such an integral representation; χ is simply an *operator-valued function of time acting on one-body potentials*.

The key observation which allows one to bring to bear TDDFT is now the following: with the restriction (4) to one-body perturbing potentials and one-body observables, the expectation value $\langle V_{\mathscr{O}} \rangle_t$ only depends on the electronic density ρ^{Ψ} at time t,

$$\rho^{\Psi}(t, r) = N \int_{\mathbb{R}^{3(N-1)}} |\Psi(t, r, r_2, \ldots, r_N)|^2 \, dr_2 \ldots dr_N. \tag{6}$$

In the following we write $\rho^{\Psi}(t)$ for the function $\rho^{\Psi}(t, \cdot) \colon \mathbb{R}^3 \to \mathbb{R}$. We have

$$\langle V_{\mathscr{O}} \rangle_t = \langle v_{\mathscr{O}}, \rho^{\Psi}(t) \rangle = \langle v_{\mathscr{O}}, \rho^{\Psi}(0) \rangle + \varepsilon \langle v_{\mathscr{O}}, (\chi v_{\mathscr{P}} \star f)(t) \rangle + \mathcal{O}(\varepsilon^2), \tag{7}$$

so by identification, χ gives the variation of the electronic density to the first order in ε,

$$\rho^{\Psi}(t) = \rho^{\Psi}(0) + \varepsilon(\chi v_{\mathscr{P}} \star f)(t) + \mathcal{O}(\varepsilon^2), \tag{8}$$

where throughout this paper $a \star f$ denotes convolution in time,

$$(a \star f)(t) = \int_0^t a(t-s) f(s) \, ds.$$

If it is possible to efficiently approximate the density evolution, and hence the density–density response function χ, then we have a way to obtain the variation of the expectation value $\langle V_{\mathscr{O}} \rangle_t$ in the linear response regime. Such approximations are provided by TDDFT. In the next section we only introduce the simplest – and commonly used – such approximation, referring to Appendix A for more details and references.

1.2. Time-dependent density functional theory

TDDFT aims to reproduce or approximate the evolution of the electronic density ρ^{Ψ}, which is governed by the many-body Hamiltonian H, by the evolution of the density of a noninteracting system. More precisely, the electronic density ρ^{Ψ} is approximated by the electronic density ρ^{Φ}, where Φ is the solution to

$$\begin{cases} i \, \partial_t \Phi(t) = H_{\text{eff}}(t) \Phi(t), & t > 0, \\ \Phi(0) = \Phi_0, \end{cases} \tag{9}$$

for some suitable effective noninteracting Hamiltonian H_{eff}. The initial condition Φ_0 is a Slater determinant whose density ρ^{Φ_0} approximates the electronic density $\rho^{\Psi}(0)$ of the exact ground state. In practice it is taken to be the Kohn–Sham determinant, i.e. the ground state of the Hamiltonian (12). In this paper we focus on the physical case of the Coulomb

interaction in (2), $w = \frac{1}{|\cdot|}$, and the *random phase approximation (RPA)*, which corresponds to the effective Hamiltonian

$$H_{\text{eff}}(t) = -\frac{1}{2}\Delta + \sum_{i=1}^{N} v(r_i) + v_{\text{xc}}^{\text{static}}[\rho^{\Phi}(0)](r_i) + \rho^{\Phi}(t) * \frac{1}{|\cdot|}(r_i) + \varepsilon f(t) v_{\mathscr{P}}(r_i). \quad (10)$$

Here, Φ is the solution to (9), ρ^{Φ} is its density (equation (6) with Ψ replaced by Φ), $v_{\text{xc}}[\rho^{\Phi}(0)]$ is the static Kohn–Sham exchange-correlation potential of the initial density, and $\frac{1}{|\cdot|} * \cdot$ is the Hartree operator, i.e. the convolution with the Coulomb potential. Thus, in the RPA, the Hartree potential $\rho^{\Phi}(t) * \frac{1}{|\cdot|}$ (being the dominant part of the interaction) is dynamically updated, whereas the exchange-correlation potential is frozen at the initial density. Also updating it dynamically would correspond to the adiabatic local density approximation (ALDA) provided $v_{\text{xc}}[\rho]$ is given by the LDA. Since we are interested in the linear response regime, it is not necessary to solve the nonlinear system (9), (10). Instead, assuming that ρ^{Φ} has a Taylor expansion to order 1 in ε, it can be shown (see Appendix A) that the variation of $\langle V_{\mathscr{O}} \rangle_t$ is given by formula (7) with χ replaced by the solution χ^{RPA} to the following Dyson equation:

$$\chi^{\text{RPA}}(t) = \chi_0(t) + \left(\chi_0 \star \left(\frac{1}{|\cdot|} * \chi^{\text{RPA}} \right) \right)(t). \quad (11)$$

The operator-valued function χ_0 is the density–density response function of the frozen (or static) Hamiltonian

$$H_0 = -\frac{1}{2}\Delta + \sum_{i=1}^{N} v(r_i) + v_{\text{xc}}^{\text{static}}[\rho^{\Phi}(0)](r_i) + \rho^{\Phi}(0) * \frac{1}{|\cdot|}(r_i). \quad (12)$$

Equation (11) is called the Dyson equation in the RPA, and its solution is the RPA density–density response function [16, 22]. This is the simplest approximation of the density–density response function χ in TDDFT. More sophisticated approximations (see e.g. [15]) are beyond the scope of the present paper. The interested reader may consult Appendix A for a compact introduction to TDDFT and the derivation of (11), and the monographs [16, 22] for an overview of the field from a physics perspective.

Finally, we remark that interesting linear response problems arise in many different contexts. For mathematical results on the potential-to-density response of nonlinear Hartree dynamics for crystals see [4]. A mathematical study of the linear response of density and current density in TDDFT to applied electromagnetic fields in a continuum limit can be found in [8].

1.3. Main results

The main focus of the paper is to establish fundamental properties of the solution χ^{RPA} to the Dyson equation (11) (including but not limited to existence and uniqueness), under natural assumptions on the Hamiltonian H associated to the reference density–density response function χ_0.

1.3.1. Assumptions. We recall here that the rest Hamiltonian considered is

$$H = -\frac{1}{2}\Delta + \sum_{1 \le i < j \le N} w(r_i - r_j) + \sum_{i=1}^{N} v(r_i), \tag{13}$$

acting on the anti-symmetric L^2-space

$$\mathcal{H}_N = \bigwedge_{i=1}^{N} L^2(\mathbb{R}^3),$$

where $v, w \in L^2(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ are real valued. Under this condition on v and w, it is well known that the Hamiltonian H is a self-adjoint operator with domain $H^2(\mathbb{R}^{3N}) \cap \mathcal{H}_N$ which is bounded from below [18].

Throughout this paper, we shall also assume that H satisfies the following general assumption.

Assumption 1. Let $v, w \in L^2(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ be real-valued functions and H be defined as in (13). Then we assume that

(i)   the ground state energy E_0 of H is a simple isolated eigenvalue;

(ii) the electronic density ρ^{Ψ_0} of the ground state Ψ_0 is bounded;

Note that assumption (ii) is in fact a consequence of (i) for Schrödinger operators of the form (13) (see e.g. [20]). The reason we promote it to an independent assumption here is that all the results from this paper ultimately rely on these two assumptions and not on the specific form of the rest Hamiltonian H.

Under the above assumptions we note that the ionization threshold Ω, defined as

$$\Omega = \inf \sigma_{\text{ess}}(H) - E_0, \tag{14}$$

is positive. This is a simple but important observation, since most of the results discussed next concern the behavior of $\widehat{\chi^{\text{RPA}}}$ inside the interval $(-\Omega, \Omega)$.

1.3.2. Solution to the Dyson equation. We first show that the Dyson equation (11) has a unique solution in the space of strongly continuous maps from $\mathbb{R}_+$ to $\mathcal{B}(L^2(\mathbb{R}^3) + L^\infty(\mathbb{R}^3), L^1(\mathbb{R}^3) \cap L^2(\mathbb{R}^3))$, denoted here by

$$C_s(\mathbb{R}_+; \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2)).$$

This is the space of density–density response functions of Hamiltonians (2) as seen in Proposition 2.4. The proof is an application of the Banach fixed point theorem in an appropriate space.

Theorem 1.1 (Existence of the solution χ^{RPA}). *Let $\chi_0 \in C_s(\mathbb{R}_+; \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2))$. Then the following assertions are true:*

(i) *there is a unique solution $\chi^{\mathrm{RPA}} \in C_s(\mathbb{R}_+; \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2))$ to the RPA Dyson equation* (11)*;*

(ii) *the solution map*

$$\mathcal{S}^{\mathrm{RPA}} : \begin{cases} C_s(\mathbb{R}_+; \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2)) \to C_s(\mathbb{R}_+; \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2)), \\ \chi_0 \mapsto \chi^{\mathrm{RPA}}, \end{cases}$$

is a bijection.

1.3.3. Poles of the RPA density–density response function. From equations (3) and (5), it can be shown (see Section 2.3) that the Fourier transform of the density–density response function χ, defined on $\{\mathrm{Im}(z) > 0\}$ as $\int_0^\infty \chi(t) e^{izt} \, \mathrm{d}t$, is an analytic family of operators whose meromorphic extension has simple real poles. This means that for $\omega \in \mathbb{R}$ a pole of $\hat{\chi}$, there is a neighborhood of ω, an analytic family of operators defined in this neighborhood $z \mapsto K_0(z)$, and a finite-rank operator K_{-1}, such that

$$\hat{\chi}(z) = K_0(z) + \frac{K_{-1}}{z - \omega}.$$

We denote the set of poles of $\hat{\chi}$ by $\mathcal{P}(\hat{\chi})$ and define the rank of a pole ω as

$$\mathrm{rank}_\omega(\hat{\chi}) = \mathrm{rank}\, K_{-1}.$$

The main result of this paper shows that the RPA density–density response function has a similar structure. More precisely, we show that the Fourier transform of χ^{RPA} also admits a meromorphic extension whose poles are located along the real axis. We also prove that these poles are forward shifted compared to the poles of the Fourier transform of the reference density–density response function χ_0.

Note that even the existence of the Fourier transform in the upper half-plane is not clear a priori. Indeed, if we take $\chi_0(t) = K$ for any $t \geq 0$ and some bounded operator $K \in \mathcal{B}(L^2(\mathbb{R}^3) + L^\infty(\mathbb{R}^3); L^1(\mathbb{R}^3) \cap L^2(\mathbb{R}^3))$, then χ_0 is a strongly continuous map from $\mathbb{R}_+$ to $\mathcal{B}(L^2 + L^\infty, L^1 \cap L^2)$ but the solution to the Dyson equation (11) in this case is given by $e^{tKF_H} K$, whose Fourier transform diverges in $\{\mathrm{Im}(z) < \sup \sigma(KF_H)\}$.

Theorem 1.2 (Poles of $\widehat{\chi^{\mathrm{RPA}}}$). *Let χ_0 be the density–density response function (see the definition in Proposition* 2.3*) of a Hamiltonian H satisfying Assumption* 1*. Let χ^{RPA} be the solution of the RPA Dyson equation* (11) *and $\Omega = \inf \sigma_{\mathrm{ess}}(H) - E_0$ be the ionization threshold of H. Let $\mathcal{D}_\Omega$ be the set*

$$\mathcal{D}_\Omega = \mathbb{C} \setminus ((-\infty, -\Omega] \cup [\Omega, \infty)).$$

Then the following holds:

(i) *(Meromorphic) The Fourier transform $\widehat{\chi^{\mathrm{RPA}}} : \mathcal{D}_\Omega \to \mathcal{B}(L^2(\mathbb{R}^3) + L^\infty(\mathbb{R}^3), L^1(\mathbb{R}^3) \cap L^2(\mathbb{R}^3))$ is a meromorphic family of operators with simple poles contained in $(-\Omega, \Omega)$;*

(ii) (Forward shift of poles) *Let $\mathcal{P}(\hat{\chi}_0)$ be the set of poles of $\hat{\chi}_0$. Then the poles of $\widehat{\chi^{\mathrm{RPA}}}$ are forward shifted with respect to the poles of $\hat{\chi}_0$ in the sense that, for any $0 < \omega < \Omega$,*

$$\sum_{\substack{\tilde{\omega} \in \mathcal{P}(\widehat{\chi^{\mathrm{RPA}}}) \\ |\tilde{\omega}| < \omega}} \mathrm{rank}_{\tilde{\omega}}(\widehat{\chi^{\mathrm{RPA}}}) \leq \sum_{\substack{\tilde{\omega} \in \mathcal{P}(\hat{\chi}_0) \\ |\tilde{\omega}| < \omega}} \mathrm{rank}_{\tilde{\omega}}(\hat{\chi}_0).$$

Remark 1.3. While the theorem is insensitive to the precise form of the Hamiltonian H governing the reference density–density response function χ_0, the standard choice given in equation (12) is of particular physical interest. In this case, the poles are located precisely at the spectral gaps $\epsilon_a - \epsilon_j$ between occupied and unoccupied eigenvalues of the (one-body) Kohn–Sham Hamiltonian

$$h_{\mathrm{KS}} = -\frac{1}{2}\Delta + v + v_{\mathrm{xc}}[\rho^{\Phi}(0)] + \rho^{\Phi}(0) * \frac{1}{|\cdot|}$$

of time-independent density functional theory (see Appendix B). Thus statement (ii) implies that the Kohn–Sham spectral gaps (accounting for multiplicities) always under-estimate the excitation frequencies predicted by the RPA. This can be taken as a mathematical explanation of the empirical fact that the Kohn–Sham gaps are commonly too low compared to the experimental excitation frequencies (see e.g. [24]).

As a last result, we give a rigorous way to find the poles of $\widehat{\chi^{\mathrm{RPA}}}$, and compute its rank, by solving an eigenvalue problem. This rigorously justifies the standard approach in the quantum chemistry community to find the poles of $\widehat{\chi^{\mathrm{RPA}}}$, as long as they lie outside the set of poles of $\hat{\chi}_0$. For the mutual poles of $\hat{\chi}_0$ and $\widehat{\chi^{\mathrm{RPA}}}$, the rank can also be computed by solving a similar eigenvalue problem in a reduced space. More precisely, we have the following criteria:

Theorem 1.4 (Characterization of the poles of $\widehat{\chi^{\mathrm{RPA}}}$). *Let $|\omega| < \Omega$ be a pole of $\widehat{\chi^{\mathrm{RPA}}}$, and let F_H be the Hartree operator $f \mapsto \frac{1}{|\cdot|} * f$. Then the following hold:*

(i) *If ω is not a pole of $\hat{\chi}_0$, then the rank of ω as a pole of $\widehat{\chi^{\mathrm{RPA}}}$ is given by the number of linearly independent solutions in $L^2(\mathbb{R}^3)$ of the equation*

$$F_H^{1/2} \hat{\chi}_0(\omega) F_H^{1/2} f = f. \tag{15}$$

(ii) *If ω is a pole of $\hat{\chi}_0$, then the rank of ω as a pole of $\widehat{\chi^{\mathrm{RPA}}}$ is given by the number of linearly independent solutions in $L^2(\mathbb{R}^3)$ of*

$$P_V^{\perp} F_H^{1/2} \hat{\chi}_0(\omega) F_H^{1/2} P_V^{\perp} f = f,$$

where $F_H^{1/2}$ is the square root of F_H given up to a normalization constant by convolution against $\frac{1}{|\cdot|^2}$, and $P_V^{\perp}$ is the orthogonal projection on the orthogonal complement of $V = F_H^{1/2}(\ker(H - E_0 + |\omega|))$.

Remark 1.5. Note that for any solution f of (15), the nonzero function $g = \hat{\chi}_0(\omega) F_H^{1/2} f$ $\in L^1(\mathbb{R}^3) \cap L^2(\mathbb{R}^3)$ satisfies the equation $\hat{\chi}_0(\omega) F_H g = g$. Likewise, a solution g of the latter yields a solution of the former by taking $f = F_H^{1/2} g \in L^2(\mathbb{R}^3)$. In particular, solving (15) is equivalent to solving $g = \hat{\chi}_0(\omega) F_H g$, which is the starting point of the Casida formalism in TDDFT [14, Section 3.8].

Strategy of the proof. The proofs of Theorems 1.2 and 1.4 consist of two main steps. The first and most involved step is a detailed spectral analysis of the symmetrized operator $\hat{\chi}_s = F_H^{1/2} \hat{\chi} F_H^{1/2}$. This analysis leads to the characterization of the poles of $(1 - \hat{\chi}_s)^{-1}$ in Propositions 4.2 and 4.3 below. With these propositions, the second step in our proof is to show that $\widehat{\chi^{\text{RPA}}}$ is a meromorphic family of operators with simple real poles, and then prove the identity $\text{rank}_\omega(\widehat{\chi^{\text{RPA}}}) = \text{rank}_\omega((1 - \hat{\chi}_s)^{-1})$ for any $\omega \in (-\Omega, \Omega)$. The key properties of F_H used in our proofs are its positivity and its L^p-mapping properties, given by the Hardy–Littlewood–Sobolev inequality. In particular, we expect that the results here can be extended to other types of adiabatic approximations used in TDDFT.

Structure of the paper. We start by introducing some notation in the next paragraph. In Section 2 we introduce the (exact) density–density response function χ of a general Hamiltonian and relate it to the celebrated Kubo formula from linear response theory. We also derive some L^p smoothing properties of χ (Lemma 2.4) and give the formula of its Fourier transform, which can be viewed as a meromorphic family of operators. In Section 3 we prove the existence and uniqueness of solutions to the RPA-Dyson equation (11) in the setting of Theorem 1.1. In Section 4 we study the symmetrized operator $\hat{\chi}_s = F_H^{1/2} \hat{\chi}_0 F_H^{1/2}$ and characterize the poles of $(1 - \hat{\chi}_s)^{-1}$ in Propositions 4.2 and 4.3. We then use these propositions in Section 5 to prove Theorems 1.2 and 1.4.

Notation

The set $\mathbb{R}_+ = [0, \infty)$ denotes the set of nonnegative real numbers. For A and B nonnegative scalar quantities, $A \lesssim B$ means that there is an irrelevant positive constant C such that $A \leq CB$. We use the following convention for the Fourier transform of functions $f: \mathbb{R} \to F$ where F is a Banach space:

$$\hat{f}(\omega) = \int_{\mathbb{R}} f(t) e^{it\omega} \, dt.$$

Let F, G be Banach spaces. We will denote their respective norms by $\|\cdot\|_F$ and $\|\cdot\|_G$. Moreover, we denote the set of linear continuous operators from F to G by $\mathcal{B}(F, G)$. If $F = G$, we simply use $\mathcal{B}(F)$. The operator norm on $\mathcal{B}(F, G)$ is denoted by

$$\|T\|_{F,G} = \sup_{\substack{g \in G \\ \|g\|_G = 1}} \|Tg\|_F.$$

Whenever it is clear from the context to which operator space T belongs, we shall use only $\|T\|$ for the operator norm. For an operator $T: F \to G$ on Banach spaces F, G, we

denote its kernel and range by $\ker T \subset F$ and $\operatorname{ran} T \subset G$. We also use $\operatorname{rank} T = \dim \operatorname{ran} T$ for the rank of T. For $1 \leq p \leq \infty$, $L^p(\mathbb{R}^3)$ (or just L^p) denotes the standard L^p spaces with respect to Lebesgue measure. We use the notation

$$\langle f, g \rangle = \int_{\mathbb{R}^n} \overline{f(r)} g(r) \, \mathrm{d}r$$

for the standard inner product on $L^2(\mathbb{R}^n)$. We also use $L^p(\mathbb{R}^n) + L^q(\mathbb{R}^n)$ and $L^p(\mathbb{R}^n) \cap L^q(\mathbb{R}^n)$ for the Banach spaces of measurable functions with finite norms

$$\|f\|_{L^p + L^q} = \inf_{f = f_p + f_q} \{\|f_p\|_{L^p} + \|f_q\|_{L^q}\},$$

$$\|f\|_{L^p \cap L^q} = \max\{\|f\|_{L^p}, \|f\|_{L^q}\}.$$

Note that if p and q are conjugate exponents, that is to say, $p^{-1} + q^{-1} = 1$, $L^p + L^q$ is the dual of $L^p \cap L^q$. For a projection $P \in \mathcal{B}(F)$ on a Banach space F, i.e. $P^2 = P$, we say that an operator $B \in \mathcal{B}(F)$ is invertible with respect to P if $PBP = B$ and there exists an operator $B^{-1} \in \mathcal{B}(F)$ such that

$$PB^{-1}P = B^{-1} \quad \text{and} \quad B^{-1}B = BB^{-1} = P.$$

Note that the inverse B^{-1} is unique.

2. The ground-state density–density response function

In this section we recall the basics of linear response theory and give a derivation of the density–density response function. We then highlight a few properties of this operator-valued function and give a representation of its Fourier transform.

2.1. Derivation and Kubo formula

Let us start with the definition of the linear response function and its connection to the first-order variation in the dynamics with respect to some perturbation.

Let H be the static Hamiltonian defined in (13) and consider the time-dependent family of self-adjoint operators $H(t)$,

$$H(t) = H + \varepsilon \theta(t) f(t) V_{\mathcal{P}}, \tag{16}$$

where the perturbing potential $V_{\mathcal{P}}$ is a bounded multiplication operator $V_{\mathcal{P}} \colon \mathcal{H}_N \to \mathcal{H}_N$, the function $f \in L^\infty(\mathbb{R})$, $\varepsilon \in \mathbb{R}$, and θ is the Heaviside function

$$\theta(t) = \begin{cases} 0 & \text{if } t < 0, \\ 1 & \text{otherwise.} \end{cases}$$

Next suppose that $V_{\mathcal{O}}\colon \mathcal{H}_N \to \mathcal{H}_N$ is an observable of interest. We are interested in the expectation value $\langle V_{\mathcal{O}}\rangle_t := \langle \Psi(t), V_{\mathcal{O}}\Psi(t)\rangle$ for small time t where Ψ is the solution of the time-dependent Schrödinger equation

$$\begin{cases} i\,\partial_t \Psi(t) = H(t)\Psi(t), & t > 0, \\ \Psi(0) = \Psi_0, \end{cases} \tag{17}$$

with Ψ_0 being the ground-state wave function of H.

Proposition 2.1. *Let $H(t)$ be the family of self-adjoint operators defined in* (16). *Let $\Psi(t)$ be the solution of* (17) *and $V_{\mathcal{O}}\colon \mathcal{H}_N \to \mathcal{H}_N$ be a bounded multiplication operator. Then $\langle V_{\mathcal{O}}\rangle_t = \langle \Psi(t), V_{\mathcal{O}}\Psi(t)\rangle$ has the expansion*

$$\langle V_{\mathcal{O}}\rangle_t - \langle V_{\mathcal{O}}\rangle_0 = i\varepsilon \int_{-\infty}^{\infty} \theta(t - t')f(t')\langle \Psi_0, [V_{\mathcal{P}}, (V_{\mathcal{O}})_I(t - t')]\Psi_0\rangle \, dt' + \mathcal{O}(\varepsilon^2), \tag{18}$$

where $(V_{\mathcal{O}})_I(\tau) = e^{i\tau H}V_{\mathcal{O}}e^{-i\tau H}$ and $[A, B] = AB - BA$ denotes the commutator.

Proof. Let $H_1(t) = \varepsilon\theta(t)f(t)V_{\mathcal{P}}$. Under the assumption on $V_{\mathcal{P}}$ and f, it is clear that $t \mapsto H_1(t) \in \mathcal{B}(\mathcal{H}_N, \mathcal{H}_N)$ is uniformly bounded. Hence, by a standard evolution equations argument, the solution to the time-dependent Schrodinger equation exists, is unique, and satisfies the Duhamel formula

$$\Psi(t) = e^{-itH}\Psi_0 - i\int_0^t e^{-i(t-s)H}H_1(s)\Psi(s)\,ds. \tag{19}$$

In particular, iterating (19) twice yields

$$\Psi(t) = e^{-itE_0}\Psi_0 - i\varepsilon \int_0^t f(s)\theta(s)e^{-i(t-s)H}V_{\mathcal{P}}e^{-isH}\Psi_0\,ds + \mathcal{O}(\varepsilon^2). \tag{20}$$

Hence, plugging (20) into the definition of $\langle V_{\mathcal{O}}\rangle_t$ completes the proof. $\blacksquare$

If the perturbation operator $V_{\mathcal{P}}$ and the observable $V_{\mathcal{O}}$ are given by one-body potentials

$$V_{\mathcal{P}} = \sum_{k=1}^{N} v_{\mathcal{P}}(r_k), \quad V_{\mathcal{O}} = \sum_{k=1}^{N} v_{\mathcal{O}}(r_k),$$

with $v_{\mathcal{O}}$ and $v_{\mathcal{P}}$ real-valued bounded functions, then the integrand in (18) is bilinear in the potentials, providing the following bilinear form on $L^{\infty}(\mathbb{R}^3) \times L^{\infty}(\mathbb{R}^3)$:

$$\langle v_{\mathcal{O}}, \chi(\tau)v_{\mathcal{P}}\rangle = i\theta(\tau)\left\langle \Psi_0, \left[\sum_{k=1}^{N} v_{\mathcal{P}}(r_k), \left(\sum_{k=1}^{N} v_{\mathcal{O}}(r_k)\right)_I(\tau)\right]\Psi_0\right\rangle, \tag{21}$$

where the inner product on the left is the L^2 inner product. The operator-valued function $\tau \mapsto \chi(\tau)$ defined by equation (21) is the *density–density response function* associated with the Hamiltonian H.

The reason behind this definition is the celebrated Kubo formula below, which gives the first-order variation of $\langle V_{\mathcal{O}}\rangle_t$ caused by $V_{\mathcal{P}}$, as a convolution against $\langle v_{\mathcal{O}}, \chi(t)v_{\mathcal{P}}\rangle$.

Corollary 2.2 (Kubo formula). *Let χ be as defined in* (21). *Then*

$$\langle V_{\mathcal{O}} \rangle_t - \langle V_{\mathcal{O}} \rangle_0 = \varepsilon \int_0^\infty \langle v_{\mathcal{O}}, \chi(t - t') v_{\mathcal{P}} \rangle f(t') \, \mathrm{d}t' + \mathcal{O}(\varepsilon^2).$$

2.2. Regularity of the density–density response function

We now give an alternative representation of the density–density response function of H and derive a few L^p-mapping properties that will be useful in the next sections.

Proposition 2.3. *Let H be a Hamiltonian satisfying Assumption* 1. *Let E_0 be the lowest eigenvalue of H, and Ψ_0 an associated normalized eigenfunction. Define $S\colon \mathcal{H}_N \to L^1(\mathbb{R}^3)$ to be the mapping from a many-body wave function Φ to the diagonal of the mixed one-body reduced density matrix γ^{Φ, Ψ_0}, that is to say,*

$$(S\Phi)(r) = N \int_{(\mathbb{R}^3)^{N-1}} \Phi(r, r_2, \ldots, r_N) \overline{\Psi_0(r, r_2, \ldots, r_N)} \, \mathrm{d}r_2 \ldots \mathrm{d}r_N. \tag{22}$$

Let χ be the density–density response function of H (as defined in (21)*). Then for any $f, g \in L^\infty(\mathbb{R}^3)$ real valued, one has*

$$\langle f, \chi(t)g \rangle = \langle f, 2\theta(t) S \sin(t(E_0 - H)) S^* g \rangle,$$

where $S^*\colon L^\infty(\mathbb{R}^3) \to \mathcal{H}_N$ is the adjoint of S given by

$$(S^* v)(r_1, \ldots, r_N) = \sum_{k=1}^N v(r_k) \Psi_0(r_1, \ldots, r_N). \tag{23}$$

Proof. First note that the Hamiltonian H commutes with complex conjugation, i.e.

$$\overline{H\Phi}(r_1, \ldots, r_N) = H \overline{\Phi}(r_1, \ldots, r_N).$$

Hence, the projection-valued (spectral) measure P_λ^H associated to H also commutes with complex conjugation, and therefore

$$\overline{(e^{itH}\Phi)}(r_1, \ldots, r_N) = (e^{-itH}\Phi)(r_1, \ldots, r_N)$$

for any real-valued wave function $\Phi \in \mathcal{H}_N$. Moreover, due to the uniqueness assumption on the ground state of H, we can take Ψ_0 to be real valued. Therefore, using the identity $e^{itH_N}\Psi_0 = e^{itE_0}\Psi_0$ we conclude that

$$\begin{aligned}
\langle f, \chi(t)g \rangle &= i\theta(t)(\langle S^* f, e^{it(H-E_0)} S^* g \rangle - \langle S^* g, e^{-it(H-E_0)} S^* f \rangle) \\
&= i\theta(t)(\langle S^* f, e^{it(H-E_0)} S^* g \rangle - \langle S^* f, \overline{e^{it(H-E_0)} S^* g} \rangle) \\
&= -2\theta(t)\langle S^* f, \sin(t(H - E_0)) S^* g \rangle = \langle f, \chi_0(t)g \rangle,
\end{aligned}$$

for any real-valued functions $f, g \in L^\infty(\mathbb{R}^3)$. ∎

Using the boundedness of the electronic density ρ^{Ψ_0}, we can show that χ has more regularity (in terms of L^p spaces) than simply mapping L^∞ to L^1.

Proposition 2.4 (L^p-regularity of χ). *Let S and S^* be defined by* (22) *and* (23)*, for some H satisfying Assumption* 1*. Then $S \in \mathcal{B}(\mathcal{H}_N, L^1(\mathbb{R}^3) \cap L^2(\mathbb{R}^3))$ and $S^* \in \mathcal{B}(L^2(\mathbb{R}^3) + L^\infty(\mathbb{R}^3), \mathcal{H}_N)$. In particular, $t \mapsto \chi(t)$ is a strongly continuous family of operators in $\mathcal{B}(L^2(\mathbb{R}^3) + L^\infty(\mathbb{R}^3), L^1(\mathbb{R}^3) \cap L^2(\mathbb{R}^3))$, and the map $t \mapsto \|\chi(t)\|_{L^2+L^\infty, L^1\cap L^2}$ is uniformly bounded in $\mathbb{R}_+$.*

Proof. From the boundedness of ρ^{Ψ_0} and the Cauchy–Schwarz inequality we have

$$\int_{\mathbb{R}^3} |S\Phi(r)|^2 \, dr \le N \int_{\mathbb{R}^3} \rho^{\Psi_0}(r) \int_{\mathbb{R}^{3(N-1)}} |\Phi(r, r_2, \ldots, r_N)|^2 \, dr_2 \ldots dr_N \, dr$$
$$\le N \|\rho^{\Psi_0}\|_{L^\infty} \|\Phi\|_{L^2(\mathbb{R}^{3N})},$$

and

$$\int_{\mathbb{R}^3} |S\Phi(r)| \, dr \le N \int_{(\mathbb{R}^3)^N} |\Psi_0(r, r_2, \ldots, r_N) \Phi(r, r_2, \ldots, r_N)| \, dr_2 \ldots dr_N$$
$$\le N \|\Psi_0\|_{L^2(\mathbb{R}^{3N})} \|\Phi\|_{L^2(\mathbb{R}^{3N})} = N \|\Phi\|_{L^2(\mathbb{R}^{3N})}.$$

Hence, S maps $\mathcal{H}_N$ to $L^1(\mathbb{R}^3) \cap L^2(\mathbb{R}^3)$. Since S^* is the adjoint of S, S^* is bounded from $(L^1(\mathbb{R}^3) \cap L^2(\mathbb{R}^3))^* = L^2(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ to $(\mathcal{H}_N)^* = \mathcal{H}_N$ (where we use the Riesz representation for the second identification). The properties of χ now follow because $\sin(t(H - E_0))$ is strongly continuous in $\mathcal{B}(\mathcal{H}_N, \mathcal{H}_N)$ and goes to 0 strongly as $t \to 0$. ∎

2.3. Fourier transform of the density–density response function and its poles

Here we give a representation of the Fourier transform of the density–density response function χ in terms of the resolvent of H. This can be viewed as a mathematically rigorous version of the celebrated Lehmann representation, to which it reduces under the – in the physics literature tacitly made but for the physical Hamiltonian (13) incorrect – assumption of purely discrete spectrum. We also introduce the definition of a meromorphic family of operators with poles of finite rank, borrowed from [7, Appendix C], and show that the poles of $\hat{\chi}$ are located at the spectrum of H. Let us start with the Fourier transform of χ, defined on the upper half-plane $\{\text{Im}(z) > 0\}$ by

$$\hat{\chi}(z) = \int_0^\infty \chi(t) e^{izt} \, dt, \tag{24}$$

where we have used that $\chi(t) = 0$ for $t < 0$.

Proposition 2.5 (Fourier transform of the density–density response function). *Let χ be the density–density response function defined in* (21) *for some H satisfying Assumption* 1*. Then the Fourier transform of χ is given by*

$$\hat{\chi}(z) = S(1 - P_{E_0}^H)((E_0 - z - H)^{-1} + (E_0 + z - H)^{-1})(1 - P_{E_0}^H)S^*$$

for any $\text{Im}(z) > 0$, *where the operators* S *and* S^* *are defined in* (22), (23), E_0 *is the ground state of* H, *and* $P_{E_0}^H$ *is the orthogonal projection onto the space spanned by* Ψ_0. *Moreover, the Fourier transform of* χ *along the real line is the tempered distribution given by*

$$\hat{\chi}(\omega) = \lim_{\eta \to 0^+} S(1 - P_{E_0}^H)((E_0 - \omega - i\eta - H)^{-1} + (E_0 + \omega + i\eta - H)^{-1})$$
$$\times (1 - P_{E_0}^H)S^*, \tag{25}$$

where the limit exists in the distributional sense.

Proof. Since H is self-adjoint, from Proposition 2.3 and the spectral theorem we find that

$$\int_0^\infty \chi(t)e^{i(\omega + i\eta)t}\,dt = S\int_0^\infty \int_{E_0}^\infty 2\sin(t(E_0 - \lambda))e^{i(\omega + i\eta)t}\,dP_\lambda^H\,dt\,S^*$$
$$= S\int_{E_0}^\infty \frac{1}{E_0 - \omega - i\eta - \lambda} + \frac{1}{E_0 + \omega + i\eta - \lambda}\,dP_\lambda^H\,S^*$$
$$= S(1 - P_{E_0}^H)((E_0 - \omega - i\eta - H)^{-1}$$
$$+ (E_0 + \omega + i\eta - H)^{-1})(1 - P_{E_0}^H)S^*,$$

where P_λ^H is the spectral projection of H and we have used that

$$(E_0 + \omega + i\eta - H)^{-1}\Psi_0 + (E_0 - \omega - i\eta - H)^{-1}\Psi_0 = 0. \qquad \blacksquare$$

Next, we make formula (25) more explicit in terms of the spectrum of H.

Proposition 2.6 (Rigorous Lehmann representation). *Under the assumptions of Proposition* 2.5, *for* $\text{Im}(z) > 0$ *we have*

$$\hat{\chi}(z) = \sum_{E_j \in \sigma_d(H)\setminus\{E_0\}} SP_{E_j}^H\left(\frac{1}{E_0 - z - E_j} + \frac{1}{E_0 + z - E_j}\right)P_{E_j}^H S^*$$
$$+ S\int_{\sigma_{\text{ess}}(H)} \frac{1}{E_0 - z - \lambda} + \frac{1}{E_0 + z - \lambda}\,dP_\lambda^H S^*, \tag{26}$$

where P_λ^H *is the spectral projection of* H, *and* σ_d *and* σ_{ess} *denote, respectively, the discrete and the essential spectrum of* H.

Proof. This follows immediately from Proposition 2.5 and the spectral theorem for self-adjoint operators. $\qquad \blacksquare$

Remark 2.7 (Lehmann representation in the physics literature). When H has purely discrete spectrum, as happens for example when v is a trapping potential, formula (26) simplifies further. Let $\{\Psi_j\}_{j=0}^\infty$ be an orthonormal basis consisting of eigenstates of H, corresponding to eigenvalues E_j, and use that

$$(1 - P_{E_0}^H)\Phi = \sum_{j=1}^\infty \Psi_j\langle\Psi_j, \Phi\rangle.$$

Introduce the excitation frequencies $\omega_j = E_j - E_0$, and calculate

$$\langle v_{\mathcal{O}}, \hat{\chi}(z) v_{\mathcal{P}} \rangle$$
$$= \sum_{j \geq 1} \int v_{\mathcal{O}}(r_1)(\hat{\chi}(z) v_{\mathcal{P}})(r_1) \, dr_1$$
$$= \sum_{j \geq 1} N \int v_{\mathcal{O}}(r_1) \int \overline{\Psi_0(r_1, \ldots, r_N)} \Psi_j(r_1, \ldots, r_N) \, dr_2 \ldots dr_N \, dr_1$$
$$\times \left(\frac{1}{-z - \omega_j} + \frac{1}{z - \omega_j} \right) \langle \Psi_j, S^* v_p \rangle,$$

and therefore, decomposing z into its real and imaginary parts, i.e. $z = \omega + i\eta$ ($\eta > 0$),

$$\langle v_{\mathcal{O}}, \hat{\chi}(\omega + i\eta) v_{\mathcal{P}} \rangle = \sum_{j \geq 1} \langle \Psi_0, V_{\mathcal{O}} \Psi_j \rangle \langle \Psi_j, V_{\mathcal{P}} \Psi_0 \rangle$$
$$\times \left(\frac{1}{-(\omega + i\eta) - \omega_j} + \frac{1}{(\omega + i\eta) - \omega_j} \right). \tag{27}$$

This is precisely the *Lehmann representation* of the density–density response function familiar from the physics literature. This representation beautifully reveals how a frequency-dependent perturbation couples to the excitation spectrum of the system.

Note, however, that this representation, unlike ours above (equation (26)), is not strictly speaking applicable to the standard atomic and molecular Hamiltonians (equation (13) with $v(r) = -\sum_{\alpha=1}^{M} Z_\alpha / |r - R_\alpha|$) which contain continuous spectrum, as it misses the integral term in (26).

Let us now recall the definition of a meromorphic family of operators as defined in [7, Appendix C].

Definition 2.8 (Meromorphic family of operators). Let $\mathcal{D} \subset \mathbb{C}$ be an open set and E, F be Banach spaces. We say that $K \colon \mathcal{D} \to \mathcal{B}(E, F)$ is a meromorphic family of operators if in a neighborhood of any $z_0 \in \mathcal{D}$, there exist finite rank operators $K_{-j} \in \mathcal{B}(E, F)$, for $1 \leq j \leq k$, such that

$$K(z) = K_0(z) + \sum_{j=1}^{k} \frac{K_{-j}}{(z - z_0)^j},$$

where $K_0(z)$ is holomorphic near z_0. If $k = 1$, we say that z_0 is a simple pole and define its rank as $\operatorname{rank}_{z_0}(K) = \operatorname{rank} K_{-1}$.

Then we can relate the definition above with the representation in Proposition 2.5.

Proposition 2.9 (Poles of $\hat{\chi}$). *Let χ be the density–density response function defined in Proposition 2.3 for some Hamiltonian H satisfying Assumption 1. Let $D_\Omega \subset \mathbb{C}$ be the set*

$$\mathcal{D}_\Omega = \mathbb{C} \setminus ((-\infty, -\Omega] \cup [\Omega, +\infty)).$$

Then $\hat{\chi}\colon \mathcal{D}_\Omega \to \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2)$ is a meromorphic family of operators with simple poles contained in $(-\Omega, \Omega)$. Moreover, the set of poles of $\hat{\chi}$ is

$$\mathcal{P}(\hat{\chi}) = \{\omega \in \mathbb{R} \colon E_0 + |\omega| \in \sigma(H) \setminus E_0 \text{ and } SP^H_{E_0+|\omega|} \neq 0\}, \tag{28}$$

and the rank of a pole $\omega \in \mathcal{P}(\hat{\chi})$ is given by

$$\operatorname{rank}_\omega(\hat{\chi}) = \operatorname{rank} SP^H_{E_0+|\omega|},$$

where $P^H_{E_0+|\omega|}$ is the spectral projection of H onto the eigenspace $\ker(H - E_0 - |\omega|)$.

Proof. For $\operatorname{Im}(z) > 0$, we start from the representation of $\hat{\chi}(z)$ in Proposition 2.6. As $\Omega > 0$, we can extend $\hat{\chi}(z)$ analytically to the lower half-plane $\operatorname{Im}(z) < 0$ and we directly obtain that it is a meromorphic family of operators with simple real poles with the characterization (28). For the statement on the rank of the poles, note that from (26), $\operatorname{rank}_\omega(\hat{\chi}) = \operatorname{rank}(SP^H_{E_0+\omega}S^*)$. Moreover,

$$\langle f, SP^H_{E_0+\omega}S^*f \rangle = \langle P^H_{E_0+\omega}S^*f, P^H_{E_0+\omega}S^*f \rangle = \|P^H_{E_0+\omega}S^*f\|^2_{L^2(\mathbb{R}^{3N})},$$

for any $f \in L^2(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$, and so we have $\operatorname{rank}(SP^H_{E_0+\omega}S^*) \geq \operatorname{rank}(P^H_{E_0+\omega}S^*)$. But $\operatorname{rank}(P^H_{E_0+\omega}S^*) = \operatorname{rank}(SP^H_{E_0+\omega})$, and we already have that $\operatorname{rank}(SP^H_{E_0+\omega}S^*) \leq \operatorname{rank}(SP^H_{E_0+\omega})$, hence $\operatorname{rank}_\omega(\hat{\chi}) = \operatorname{rank}(SP^H_{E_0+\omega})$. ∎

Remark 2.10. If the Hamiltonian H has purely discrete spectrum (for instance when v is a trapping potential), $\mathcal{D}_\Omega = \mathbb{C}$ and $\mathcal{P}(\hat{\chi})$ is the whole set of singular points of the Fourier transform $\hat{\chi}$. However, past the ionization threshold (see (14)) it is not clear how singular $\hat{\chi}$ is. For instance, under suitable assumptions on v and w one can use the celebrated *limiting absorption principle* [2,3,21] to show that $\hat{\chi}$ is continuous – or even differentiable – above the ionization threshold (and away from embedded eigenvalues).

3. The RPA Dyson equation

The goal of this section is to prove Theorem 1.1. We start with existence and uniqueness, and then prove the bijection property. To shorten the notation, for any $T > 0$ we define

$$\|\chi\|_T := \operatorname*{ess\,sup}_{t \in (0,T]} \|\chi(t)\|_{L^2+L^\infty, L^1 \cap L^2}, \tag{29}$$

where $\chi \in L^\infty([0, T]; \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2))$.

3.1. Well-posedness of the Dyson equation

We now turn to the well-posedness of the RPA-Dyson equation

$$\chi^{\mathrm{RPA}}(t) = \chi_0(t) + \int_0^t \chi_0(t-s) F_H \chi^{\mathrm{RPA}}(s) \, \mathrm{d}s,$$

where F_H is the Hartree operator defined as the convolution with $\frac{1}{|\cdot|}$. Here, the reference response function χ_0 can be a general operator-valued function of time, only required to satisfy mild regularity conditions.

The starting point is to show that the convolution map

$$(\chi_0, \chi) \mapsto \mathcal{C}(\chi_0, \chi)(t) = (\chi_0 \star F_H \chi)(t) := \int_0^t \chi_0(t-s) F_H \chi(s) \, ds$$

is continuous in appropriate spaces. More precisely, we have the following lemma.

Lemma 3.1 (Continuity of the convolution map). *Let $\chi \in L^\infty((0, T], \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2))$ and $\chi_0 \in L^\infty((0, T], \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2))$. Then the function $t \mapsto \chi_0(t - s) F_H \chi(s)$ belongs to $L^\infty((0, T], \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2))$ and it holds that*

$$\|\mathcal{C}(\chi_0, \chi)\|_T \lesssim T \|\chi_0\|_T \|\chi\|_T. \tag{30}$$

Moreover, if either χ or χ_0 is strongly continuous, then so is $\mathcal{C}(\chi_0, \chi)$.

Proof. Since $F_H = 4\pi(-\frac{1}{2}\Delta)^{-1}$ and we have the continuous inclusions $L^p(\mathbb{R}^3) \subset L^2(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ and $L^1(\mathbb{R}^3) \cap L^2(\mathbb{R}^3) \subset L^q(\mathbb{R}^3)$, for $2 \leq p \leq \infty$ and $1 \leq q \leq 2$, estimate (30) follows directly from the Hardy–Littlewood–Sobolev inequality, which in $\mathbb{R}^3$ reads

$$\|I_\alpha f\|_p \leq C \|f\|_q, \quad I_\alpha = (-\Delta)^{-\frac{\alpha}{2}},$$

for $\frac{1}{q} = \frac{1}{p} + \frac{\alpha}{3}$ with $1 < p, q < \infty$. Strong continuity follows by observing that $\chi_0(t - s) F_H \chi(s)$ is strongly continuous in t and uniformly bounded (in s) in the $\mathcal{B}(L^2 + L^\infty, L^1 \cap L^2)$-operator norm. Hence, by dominated convergence we find that $\mathcal{C}(\chi_0, \chi)$ is also strongly continuous. On the other hand, if χ is strongly continuous we can use the change of variables $s \mapsto t - s$ and the same argument to show that $\mathcal{C}(\chi_0, \chi)$ is strongly continuous. $\blacksquare$

We can now use the above estimate to show the well-posedness on the space of strongly continuous $\mathcal{B}(L^2 + L^\infty, L^1 \cap L^2)$-valued functions.

Proof of item (i) *from Theorem* 1.1. First note that by inequality (30), for T sufficiently small we know that the map

$$\mathcal{C}^T(\chi_0, \cdot) \colon \begin{cases} L^\infty((0, T], \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2)) \to L^\infty((0, T], \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2)), \\ \chi \mapsto \mathcal{C}(\chi_0, \chi)(T), \end{cases}$$

is a contraction. Therefore, by the Banach fixed point theorem, there exists a unique solution $\chi_1 \in L^\infty((0, T]; \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2))$ satisfying $\chi_1 = \chi_0 + \mathcal{C}^T(\chi_0, \chi_1)$. Hence, we just need to extend this solution to the whole $\mathbb{R}_+$. For this, note that, as $\chi_0 \in L^\infty((0, 2T], \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2))$, there exists some $\delta = \delta(\|\chi_0\|_{2T}) > 0$ (with the norm defined in (29)) such that the map

$$\mathcal{C}_{T_0}^{T+\delta}(\chi_0, \chi)(t) := \int_{T_0}^t \chi_0(t-s) F_H \chi(s) \, ds$$

is also a contraction in $L^\infty([T_0 - \delta, T_0 + \delta]; \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2))$ for any $0 < T_0 \le 2T - \delta$. Hence, let $\chi_1(t)$ be the solution in $L^\infty((0, T]; \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2))$; then the map

$$\chi \mapsto \chi_0 + \int_0^T \chi_0(t - s) F_H \chi_1(s) \, \mathrm{d}s + \mathcal{C}_T^{T+\delta}(\chi_0, \chi)$$

is again a contraction and we can find a unique fixed point χ_2. Moreover, we have

$$\chi_1(t) - \chi_2(t) = \int_T^t \chi_0(t - s) F_H(\chi_1(s) - \chi_2(s)) \, \mathrm{d}s = \mathcal{C}_T^{T+\delta}(\chi_0, \chi_1 - \chi_2)(t),$$

for any $T - \delta < t \le T + \delta$. But because 0 is the unique fixed point of $\mathcal{C}_T^{T+\delta}(\chi_0, \cdot)$, we must have $\chi_1(t) = \chi_2(t)$ for a.e $T - \delta < t \le T + \delta$. We have thus extended χ_1 to $(0, T + \delta]$. To conclude, note that since δ is uniform in the interval $(0, 2T]$, we can iterate the argument to extend χ_1 to the interval $(0, 2T]$. Repeating the same steps, we can further extend the solution to the whole $\mathbb{R}_+$. Strong continuity follows from the strong continuity in Lemma 3.1. ∎

3.2. Bijection of the RPA-Dyson solution map

By virtue of Theorem 1.1 (i), we can define the solution map

$$\mathcal{S}^{\mathrm{RPA}} : \begin{cases} C_s(\mathbb{R}_+; \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2)) \to C_s(\mathbb{R}_+; \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2)), \\ \chi_0 \mapsto \chi^{\mathrm{RPA}} \in \ker\{\chi_0 + \mathcal{C}(\chi_0, \cdot) - \cdot\}. \end{cases}$$

To complete the proof of Theorem 1.1, we now show that $\mathcal{S}^{\mathrm{RPA}}$ is bijective in $C_s(\mathbb{R}_+; \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2))$.

Proof of item (ii) *of Theorem* 1.1. Note that, by repeating the arguments in the proof of item (i) of Theorem 1.1, for any $\chi \in C_s(\mathbb{R}_+; \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2))$, we can find a unique $\chi_0 \in C_s(\mathbb{R}_+; \mathcal{B}(L^2 + L^\infty, L^1 \cap L^2))$ satisfying $\chi_0 = \chi - \mathcal{C}(\chi_0, \chi)$. In particular, $\chi = \mathcal{S}^{\mathrm{RPA}}(\chi_0)$ is the unique solution of the RPA-Dyson equation, which implies that $\mathcal{S}^{\mathrm{RPA}}$ is surjective. Similarly, by the uniqueness of the solution χ_0 of $\chi_0 = \chi - \mathcal{C}(\chi_0, \chi)$, we also have injectivity of $\mathcal{S}^{\mathrm{RPA}}$ and the proof is complete. ∎

4. Symmetrized density–density response function

In this section we want to characterize the poles of $(1 - \hat{\chi}_s(z))^{-1}$ for operators of the form

$$\hat{\chi}_s(z) = T \left(\int_{\sigma(H)} 2 \frac{E_0 - \lambda}{(E_0 - \lambda)^2 - z^2} \, \mathrm{d}P_\lambda^H \right) T^*, \tag{31}$$

where T is a bounded operator from $\mathcal{H}_N$ to a Hilbert space $\mathcal{H}$, and P_λ^H is the projection-valued measure of a Hamiltonian satisfying Assumption 1. As already remarked at the end of the introduction, such a characterization is essential for the proof of Theorem 1.2.

We start by introducing some new notation. By virtue of Proposition 2.9, we define the relevant excitations $0 < \omega_1 < \omega_2 < \cdots$ as the set of positive poles of $\hat{\chi}_s$, i.e.

$$\{\omega_j\}_{j=1}^m = \{0 < \omega < \Omega : TP_{E_0+\omega}^H \neq 0\} = \mathcal{P}(\hat{\chi}_s) \cap (0, \Omega), \tag{32}$$

where $P_{E_0+\omega}^H$ is the spectral projection of H at the eigenvalue $E_0 + \omega$. For finite m, we set $\omega_{m+1} = \Omega$, where we recall that Ω is the ionization threshold defined in (14). We also call the intervals (ω_j, ω_{j+1}) the excitation-free intervals, and define the finite-dimensional subspaces

$$V_j := (\ker P_{E_0+\omega_j}^H T^*)^\perp = \operatorname{ran} TP_{E_0+\omega_j}^H.$$

Then by Proposition 2.9 (which also holds for $\hat{\chi}_s$ in place of $\hat{\chi}$), the rank of ω_j as a pole of $\hat{\chi}_s$ is given by $\dim V_j$.

Remark 4.1. Note that we can assume $\ker(1 - P_{E_0}^H)T^* = \{0\}$, as otherwise, we could simply set

$$\widetilde{\mathcal{H}} = (\ker(1 - P_{E_0}^H)T^*)^\perp = \overline{\operatorname{ran}(T(1 - P_{E_0}^H))} \subset \mathcal{H}$$

and consider $\hat{\chi}_s = P_{\widetilde{\mathcal{H}}} \hat{\chi}_s P_{\widetilde{\mathcal{H}}}$ as an operator in $\widetilde{\mathcal{H}}$.

The main goal of this section is then to prove the following propositions:

Proposition 4.2 (Characterization of the poles of $(1 - \hat{\chi}_s)^{-1}$). *Let $\hat{\chi}_s(z)$ be defined by (31) and $\mathcal{D}_\Omega = \mathbb{C} \setminus ((-\infty, -\Omega] \cup [\Omega, \infty))$. Then $(1 - \hat{\chi}_s)^{-1} : \mathcal{D}_\Omega \mapsto \mathcal{B}(\mathcal{H}, \mathcal{H})$ is a meromorphic family of operators with simple real poles with rank given by*

$$\operatorname{rank}_\omega((1 - \hat{\chi}_s)^{-1}) = \begin{cases} \dim \ker(1 - P_{V_j}^\perp \hat{\chi}_s(\omega_j) P_{V_j}^\perp) & \text{if } \omega = \omega_j \text{ for some } j \leq m, \\ \dim \ker(1 - \hat{\chi}_s(\omega)) & \text{otherwise.} \end{cases} \tag{33}$$

Moreover, for z close to ω_j we have

$$(1 - \hat{\chi}_s(z))^{-1} = (z - \omega_j)^{-1} K_{-1} + K_0 + \mathcal{O}(|z - \omega_j|), \tag{34}$$

where $K_{-1} P_{V_j} = 0$ and $\operatorname{ran} K_0 P_{V_j} \subset \operatorname{ran} K_{-1}$.

Proposition 4.3 (Forward shift of the poles of $(1 - \hat{\chi}_s)^{-1}$). *Let $\hat{\chi}_s$ be the operator defined in (31). Then the poles of $(1 - \hat{\chi}_s)^{-1}$ are forward shifted with respect to the poles of $\hat{\chi}_s$ in the sense that for any $0 < \omega < \Omega$, we have*

$$\sum_{\substack{\tilde{\omega} \in \mathcal{P}((1-\hat{\chi}_s)^{-1}) \\ |\tilde{\omega}| < \omega}} \operatorname{rank}_{\tilde{\omega}}((1 - \hat{\chi}_s)^{-1}) \leq \sum_{\substack{\tilde{\omega} \in \mathcal{P}(\hat{\chi}_s) \\ |\tilde{\omega}| < \omega}} \operatorname{rank}_{\tilde{\omega}}(\hat{\chi}_s).$$

A very crude heuristics for why such a statement might be true goes as follows. Assume that $\hat{\chi}_s$ equals $\frac{a}{z-\omega}$ with $a \neq 0$, which is the simplest function exhibiting a pole at ω. Then $(1 - \hat{\chi}_s(z))^{-1} = \frac{z-\omega}{z-(\omega+a)}$, which has a pole at $\omega + a$. Thus when $a > 0$, the pole of $(1 - \hat{\chi}_s)^{-1}$ is forward shifted with respect to that of $\hat{\chi}_s$. The plan for the rest of

the section is to first prove Proposition 4.2 and then to use it to prove Proposition 4.3. The proof of Proposition 4.2 consists of three main steps. First we show that $1 - \hat{\chi}_s(z)$ is invertible for $\text{Im}(z) \neq 0$ or $|\omega| < \omega_1$ and find an explicit estimate for the blow-up of the inverse as $\text{Im}(z) \to 0$ (Lemma 4.6). We then use this estimate to conclude that all poles in the excitation-free intervals (ω_j, ω_{j+1}) are simple and give their rank (Lemma 4.7). Finally, we isolate the singularity of $1 - \hat{\chi}_s(\omega)$ at $\omega = \omega_j$ and deal with the blowing-up and vanishing parts of $1 - \hat{\chi}_s(\omega)$ separately.

4.1. Proof of Proposition 4.2

The starting point of our analysis is the spectral decomposition

$$\hat{\chi}_s(z) = \sum_{j \leq m} \frac{2\omega_j}{z^2 - \omega_j^2} \underbrace{T P^H_{E_0+\omega_j} T^*}_{:=B_j} + \underbrace{T \left(\int_\Omega^\infty \frac{2\lambda}{z^2 - \lambda^2} \, dP^H_{E_0+\lambda} \right) T^*}_{:=B_{\text{ess}}(z)},$$

where $P^H_{E_0+\omega_j}$ is the spectral projection in the eigenspace $\ker(H - E_0 - |\omega_j|)$, and ω_j are the excitations defined in (32). Note that since

$$T P^H_{E_0+\omega_j} T^* = T P^H_{E_0+\omega_j} (T P^H_{E_0+\omega_j})^*,$$

the operators B_j are invertible with respect to the orthogonal projection P_{V_j}. This follows from the identity $\ker A = (\text{ran } A)^\perp$ valid for any symmetric operator $A \in \mathcal{B}(\mathcal{H}, \mathcal{H})$.

The first step to prove Proposition 4.2 is to show that the positive spectra of $\hat{\chi}_s$ are discrete. This follows from the following proposition.

Proposition 4.4 (Essential spectrum of $\hat{\chi}_s$). *Let $\hat{\chi}_s(z)$ be defined by* (31). *Then, for any $z \in \mathcal{D}_\Omega \setminus \mathcal{P}(\hat{\chi}_s)$, the operator $\hat{\chi}_s(z)$ satisfies*

$$\hat{\chi}_s(z) = \hat{\chi}_s(\bar{z})^* = \hat{\chi}_s(-z). \tag{35}$$

In particular, $\hat{\chi}_s(z)$ is self-adjoint for real z. Furthermore, we have

$$\sigma_{\text{ess}}(\hat{\chi}_s(\omega)) \subset (-\infty, 0],$$

for any $\omega \in (-\Omega, \Omega) \setminus \mathcal{P}(\hat{\chi}_s)$.

Proof. The symmetries in (35) are immediate from the definition in (31) and the identity $((z - H)^{-1})^* = (\bar{z} - H)^{-1}$. For the essential spectrum part, note that $\frac{2\lambda}{\omega^2 - \lambda^2} < 0$ for $|\omega| < \lambda$. This, together with the fact that $B_j = (T P^H_{E_0+\omega_j})(T P^H_{E_0+\omega_j})^*$ is nonnegative, implies that

$$\left\langle f, \frac{2\omega_j}{\omega_j^2 - \lambda^2} B_j f \right\rangle \leq 0 \quad \text{for any } 0 < \omega < \omega_j \tag{36}$$

and

$$\langle f, B_{\text{ess}}(\omega) f \rangle \leq 0 \quad \text{for any } 0 < \omega < \Omega. \tag{37}$$

In addition, since all B_j are finite rank operators, from Weyl's criterion we have

$$\sigma_{\text{ess}}(\hat{\chi}_s(\omega)) = \sigma_{\text{ess}}\left(\sum_{j \geq k} \frac{2\omega_j}{\omega^2 - \omega_j^2} B_j + B_{\text{ess}}(\omega) \right) \quad \text{for any integer } k \leq m. \qquad (38)$$

The result now follows from (36), (37), and (38) by the Rayleigh Ritz principle. ∎

4.1.1. Inverse of $1 - \hat{\chi}_s(z)$ for $\text{Im}(z) \neq 0$ or $|z| < \omega_1$. Next we want to show that $1 - \hat{\chi}_s(z)$ is invertible for any z with $\text{Im}(z) \neq 0$ or $|\text{Re}(z)| < \omega_1$. For this, we shall use the following inequality between the real and imaginary parts of $\langle f, \hat{\chi}_s(z) f \rangle$.

Lemma 4.5 (Real to imaginary ratio). *Let $\hat{\chi}_s(z)$ be defined by (31). Then for any $z = \omega + i\eta \in \mathcal{D}_\Omega \setminus ((-\Omega, -\omega_1] \cup [\omega_1, \Omega))$ and $f \in \mathcal{H}$ we have*

$$\text{Re}(\langle f, \hat{\chi}_s(z) f \rangle) \leq \max\left\{0, \frac{\omega^2 - \eta^2 - \omega_1^2}{|\omega\eta|}\right\} |\text{Im}(\langle f, \hat{\chi}_s(z) f \rangle)|. \qquad (39)$$

Proof. Let $z = \omega + i\eta$ and suppose that $\omega^2 - \eta^2 \leq \omega_1^2$. Then, since the integrand in (31) vanishes for $\lambda = E_0$, by making the translation $\lambda \mapsto \lambda - E_0$, we find that

$$\text{Re}\langle f, \hat{\chi}_s(z) f \rangle = 2 \int_{\omega_1}^{\infty} \frac{\overbrace{\lambda(\omega^2 - \eta^2 - \lambda^2)}^{\leq 0}}{|\lambda^2 + z^2|^2} \, \mathrm{d}\| P_{E_0+\lambda}^H T f \|^2 \leq 0,$$

which gives estimate (39) in this case. On the other hand, if $\omega^2 - \eta^2 > \omega_1^2$ we have

$$\text{Re}\langle f, \hat{\chi}_s(z) f \rangle \leq 2 \int_{\omega_1}^{\sqrt{\omega^2 - \eta^2}} \frac{\lambda(\omega^2 - \eta^2 - \lambda^2)}{|\lambda^2 - z^2|^2} \, \mathrm{d}\| P_{E_0+\lambda}^H T f \|^2$$

$$\leq 2 \int_{\omega_1}^{\sqrt{\omega^2 - \eta^2}} \frac{\lambda|\omega\eta|}{|\lambda^2 - z^2|^2} \left(\frac{\omega^2 - \eta^2 - \lambda^2}{|\omega\eta|} \right) \mathrm{d}\| P_{E_0+\lambda}^H T f \|^2$$

$$\leq \frac{\omega^2 - \eta^2 - \omega_1^2}{|\omega\eta|} \left| 2 \int_{\omega_1}^{\sqrt{\omega^2 - \eta^2}} \frac{\lambda\omega\eta}{|\lambda^2 - z^2|^2} \, \mathrm{d}\| P_{E_0+\lambda}^H T f \|^2 \right|$$

$$\leq \frac{\omega^2 - \eta^2 - \omega_1^2}{|\omega\eta|} |\text{Im}\langle f, \hat{\chi}_s(z) f \rangle|. \qquad ∎$$

Now we can use estimate (39) to show that $1 - \hat{\chi}_s(z)$ is invertible away from the real axis and before the first excitation ω_1. In addition, we obtain an explicit upper bound on the blow-up rate of the inverse as z approaches the real axis. This bound will be useful to show that the poles of $(1 - \hat{\chi}_s)^{-1}$ are simple.

Lemma 4.6 (Inverse away from the real axis and before ω_1). *Let $\hat{\chi}_s(z)$ be defined in (31) and $\mu_0 > 0$. Then $\mu_0 - \hat{\chi}_s(z)$ is invertible in the set $\{z = \omega + i\eta \in \mathbb{C} : \eta \neq 0 \text{ or } |\omega| \leq \omega_1\}$. Moreover, we have*

$$\|(\mu_0 - \hat{\chi}_s(\omega + i\eta))^{-1}\| \lesssim \mu_0^{-1} |z| \, |\eta|^{-1}, \qquad (40)$$

for any $z \in \mathbb{C} \setminus \mathbb{R}$.

Proof. Let $g: \mathbb{C} \setminus \{\omega_1, -\omega_1\} \to \mathbb{R} \cup \{+\infty\}$ be the function

$$g(\omega + i\eta) = \max\left\{0, \frac{\omega^2 - \eta^2 - \omega_1^2}{|\omega\eta|}\right\}, \quad (\omega, \eta) \in \mathbb{R}^2 \setminus \{(\omega_1, 0), (0, \omega_1)\}.$$

Then, for $f \neq 0$ with $\operatorname{Re}\langle f, \hat{\chi}_s(z)f\rangle \leq 0$, we have

$$\|(\mu_0 - \hat{\chi}_s(z))f\| \geq \|f\|^{-1} |\operatorname{Re}\langle f, \mu_0 - \hat{\chi}_s(z)f\rangle| \geq \mu_0 \|f\|.$$

On the other hand, by estimate (39), for any $f \in \mathcal{H}$ with $\operatorname{Re}\langle f, \hat{\chi}_s(z)f\rangle \geq 0$ and $\|f\| = 1$, we find

$$\begin{aligned}
\|(\mu_0 - \hat{\chi}_s(z))f\|^2 &\geq |\langle f, (\mu_0 - \hat{\chi}_s(z))f\rangle|^2 \\
&= (\operatorname{Re}\langle f, \mu_0 - \hat{\chi}_s(z)f\rangle)^2 + (\operatorname{Im}\langle f, \hat{\chi}_s(z)f\rangle)^2 \\
&\geq (\operatorname{Re}\langle f, \hat{\chi}_s(z)f\rangle)^2 (1 + g(z)^{-2}) - 2\mu_0 \operatorname{Re}\langle f, \hat{\chi}_s(z)f\rangle + \mu_0^2.
\end{aligned}$$

Thus, minimizing the function $\tau \mapsto \tau^2(1 + g(z)^{-2}) - 2\mu_0\tau + \mu_0^2$ we obtain

$$\|(\mu_0 - \hat{\chi}_s(z))f\| \geq \frac{\mu_0}{\sqrt{1 + g(z)^2}}\|f\|, \tag{41}$$

for any such f. We thus conclude that (41) holds for any $f \in \mathcal{H}$ with $\|f\| = 1$ (as $1 + g(z)^2 \geq 1$). Therefore, $\mu_0 - \hat{\chi}_s(z)$ is injective and the range is closed whenever $g(z) < +\infty$, which is precisely the set $\{z = \omega + i\eta : \eta \neq 0 \text{ or } |\omega| < \omega_1\}$. Moreover, since $\hat{\chi}_s(z)^* = \hat{\chi}_s(\bar{z})$ (see Proposition 4.4) and $g(z) = g(\bar{z})$, the adjoint $(\mu_0 - \hat{\chi}_s(z))^* = \mu_0 - \hat{\chi}_s(\bar{z})$ is also injective, which implies that $\mu_0 - \hat{\chi}_s(z)$ is invertible. Estimate (40) now follows from (41) and the estimate $g(z) = \max\{0, (\omega^2 - \eta^2 - \omega_1^2)/|\omega\eta|\} \leq |\omega|/|\eta|$. ∎

4.1.2. Inverse of $1 - \hat{\chi}_s(\omega)$ away from the poles of $\hat{\chi}_s$.
We now prove a lemma that will be useful to show that all poles of $(1 - \hat{\chi}_s)^{-1}$ are simple.

Lemma 4.7 (Simple poles at discrete spectrum). *Let $K: B_\epsilon(z_0) \to \mathcal{B}(\mathcal{H})$ be a holomorphic family of operators such that $K(z_0)$ is normal, 0 is an isolated point in the spectrum of $K(z_0)$, and $M = \dim \ker K(z_0) < \infty$. Suppose that there is a constant $C > 0$ such that*

$$\|K(z_0 + i\eta)f\| \geq C|\eta|\|f\|, \quad \text{for any } f \in \mathcal{H} \text{ and } \eta > 0 \text{ close to } 0. \tag{42}$$

Then $K(z)$ is invertible for $z \neq z_0$ close enough to z_0 and

$$K(z)^{-1} = \frac{K_{-1}}{z - z_0} + K_0(z),$$

where $\operatorname{rank} K_{-1} = M$ *and $K_0(z)$ is holomorphic in $B_\epsilon(z_0)$ (for some possibly smaller $\epsilon > 0$).*

Proof. First, since $0 \in \sigma_d(K(z_0))$, we know from standard perturbation theory (see Lemma C.1 in the appendix) that for any $\delta > 0$ small enough, the projection

$$Q(z) = \frac{1}{2\pi i} \oint_{\partial B_\delta(0)} (\xi - K(z))^{-1} \, \mathrm{d}\xi$$

is holomorphic for z close enough to z_0. Moreover, as $K(z_0)$ is normal, the projection $Q(z_0)$ is the orthogonal projection on $\ker K(z_0)$. Hence,

$$\lim_{z \to z_0} Q(z)K(z)Q(z) \to P_{\ker K(z_0)} K(z_0) P_{\ker K(z_0)} = 0,$$

and therefore,

$$Q(z)K(z)Q(z) = (z - z_0)K_1 + \mathcal{O}(|z - z_0|^2). \tag{43}$$

Hence, from (42) and the fact that $Q(z)$ commutes with $K(z)$, we have

$$\|K_1 v\| \gtrsim \|v\|, \quad \text{for any } v \in \operatorname{ran} Q(z_0 + i\eta) \text{ and } \eta > 0 \text{ small enough.} \tag{44}$$

This implies that $\operatorname{rank} K_1 \geq \operatorname{rank} Q(z) = \operatorname{rank} Q(z_0) = M$. Moreover, since

$$\begin{aligned}
K_1 &= \lim_{z \to z_0} \frac{1}{z - z_0} Q(z)K(z)Q(z) \\
&= \lim_{z \to z_0} Q(z)\Big(\frac{1}{z - z_0} Q(z)K(z)Q(z)\Big)Q(z) \\
&= Q(z_0)K_1 Q(z_0),
\end{aligned}$$

we conclude that $\operatorname{rank} K_1 = \operatorname{rank} Q(z_0) = M < \infty$. But since $\operatorname{ran} Q(z_0)$ is finite-dimensional and $Q(z_0)$ commutes with K_1, we see that K_1 is invertible with respect to $Q(z_0)$. This in turn implies that $Q(z)K(z)Q(z)$ is invertible with respect to $Q(z)$, for z close to z_0 excluding $z = z_0$. Indeed, this follows from (43), the fact that $Q(z)$ commutes with $Q(z)K(z)Q(z)$, and $\operatorname{rank} Q(z)K(z)Q(z) = \dim \operatorname{ran} Q(z) < \infty$. Hence, we have the decomposition

$$K(z)^{-1} = (Q(z)K(z)Q(z))^{-1} + (\widetilde{Q}(z)K(z)\widetilde{Q}(z))^{-1},$$

where $\widetilde{Q}(z) = 1 - Q(z)$, and $(Q(z)K(z)Q(z))^{-1}$, respectively $(\widetilde{Q}(z)K(z)\widetilde{Q}(z))^{-1}$ are the inverses with respect to $Q(z)$ and $\widetilde{Q}(z)$. Moreover, by the definition of $Q(z)$, we see that $0 \notin \sigma(K(z)|_{\ker \widetilde{Q}(z)})$ for any z close to z_0. Hence, the inverse $(\widetilde{Q}(z)K(z)\widetilde{Q}(z))^{-1}$ exists and is uniformly bounded (by continuity) around $z = z_0$. We are thus left with computing the pole of $(Q(z)K(z)Q(z))^{-1}$.

To compute the pole of $(Q(z)K(z)Q(z))^{-1}$, first note that from the expansion (43) and the bound (44), we find that $\|(Q(z)K(z)Q(z))^{-1}\| \lesssim |z - z_0|^{-1}$. Thus, if we multiply (43) by $(Q(z)K(z)Q(z))^{-1}$ on the left and by K_1^{-1} on the right, we obtain

$$Q(z)K_1^{-1} = (z - z_0)(Q(z)K(z)Q(z))^{-1}Q(z_0) + \mathcal{O}(|z - z_0|).$$

Hence, using that $Q(z) = Q(z_0) + \mathcal{O}(z - z_0)$ (which holds since $Q(z)$ is holomorphic), we conclude that

$$(Q(z)K(z)Q(z))^{-1} = (z - z_0)^{-1}K_1^{-1} + \mathcal{O}(1),$$

which completes the proof. Note that the remainder $\mathcal{O}(1)$ is holomorphic since the inverse of a holomorphic operator-valued function (whenever defined) is also holomorphic and any holomorphic operator which is uniformly bounded around z_0 can be extended to z_0 (by Cauchy's formula). ∎

4.1.3. Inverse of $1 - \hat{\chi}_s(\omega)$ at the poles of $\hat{\chi}_s$. We now come to the last difficulty of the proof, namely, dealing with the points $\omega = \omega_j$. The key idea here is to use the operator

$$\hat{\chi}_{s,j}(z) := P_{V_j}^\perp \hat{\chi}_s(z) P_{V_j}^\perp \in \mathcal{B}(V_j^\perp, V_j^\perp) \tag{45}$$

as a reference for separating the spectrum of $\hat{\chi}_s$. Precisely, we show here that $\mu_0 > 0$ is an eigenvalue of $\hat{\chi}_{s,j}(\omega_j)$ with multiplicity M if and only if the spectrum of $\hat{\chi}_s(z)$ close to μ_0 converges to $\{\mu_0\}$ as $z \to \omega_j$ and the associated Riesz projection has rank M. Before we show this however, we need one technical lemma. This lemma provides an asymptotic expansion for a continuous operator-valued function close to one of its poles.

Lemma 4.8 (Inverse of an operator-valued function around a pole). *Let $V \subset \mathcal{H}$ be a closed subspace, and let $B \in \mathcal{B}(\mathcal{H})$ be invertible with respect to the orthogonal projection P_V. Let $z \mapsto A(z) \in \mathcal{B}(\mathcal{H})$ be an analytic family of operators and suppose that $P_V^\perp A(z) P_V^\perp$ is invertible with respect to $P_V^\perp$, with a uniform bound. Then, for $z \in \mathbb{C}$ small enough, the operator $A(z) + z^{-1}B$ is invertible and we have*

$$(A(z) + z^{-1}B)^{-1} = (P_V^\perp A(z) P_V^\perp)^{-1} + \mathcal{O}(|z|).$$

Proof. The proof relies on a Schur complement. Writing the operator $A(z) + z^{-1}B$ by blocks, we have

$$\begin{pmatrix} z^{-1}B + P_V A(z) P_V & P_V A(z) P_V^\perp \\ P_V^\perp A(z) P_V & P_V^\perp A(z) P_V^\perp \end{pmatrix} =: \begin{pmatrix} \mathcal{A} & \mathcal{B} \\ \mathcal{C} & \mathcal{D} \end{pmatrix}.$$

The Schur complement is the operator $\mathcal{A} - \mathcal{B}\mathcal{D}^{-1}\mathcal{C}$. Using that $z\mathcal{A}$ and $\mathcal{B}\mathcal{D}^{-1}\mathcal{C}$ are uniformly bounded, the Schur complement is invertible for $|z|$ sufficiently small:

$$(\mathcal{A} - \mathcal{B}\mathcal{D}^{-1}\mathcal{C})^{-1} = \mathcal{A}^{-1} \sum_{k \geq 0} (z\mathcal{B}\mathcal{D}^{-1}\mathcal{C}(z\mathcal{A})^{-1}).$$

The result then follows from the formula for the inverse of $A(z) + z^{-1}B$ in terms of the blocks:

$$(A(z) + z^{-1}B)^{-1}$$
$$= \begin{pmatrix} (\mathcal{A} - \mathcal{B}\mathcal{D}^{-1}\mathcal{C})^{-1} & -(\mathcal{A} - \mathcal{B}\mathcal{D}^{-1}\mathcal{C})^{-1}\mathcal{B}\mathcal{D}^{-1} \\ -\mathcal{D}^{-1}\mathcal{C}(\mathcal{A} - \mathcal{B}\mathcal{D}^{-1}\mathcal{C})^{-1} & \mathcal{D}^{-1} + \mathcal{D}^{-1}\mathcal{C}(\mathcal{A} - \mathcal{B}\mathcal{D}^{-1}\mathcal{C})^{-1}\mathcal{B}\mathcal{D}^{-1} \end{pmatrix}$$
$$= \begin{pmatrix} 0 & 0 \\ 0 & (P_V^\perp A(z) P_V^\perp)^{-1}, \end{pmatrix} + \mathcal{O}(|z|). \qquad \blacksquare$$

We can now prove the previously mentioned correspondence between the spectra of $\hat{\chi}_s(z)$ and $\hat{\chi}_{s,j}(\omega_j)$ as z approaches ω_j.

Lemma 4.9 (Convergence of discrete spectra). *Let $\hat{\chi}_{s,j}(z)$ be the operator defined in (45). Then, for any $\mu_0 > 0$ and $\delta > 0$ small enough, there exists a neighborhood U_δ of ω_j such that $\partial B_\delta(\mu_0) \cap \sigma(\hat{\chi}_s(z)) = \emptyset$ for any $z \in U_\delta$ and*

$$
Q(z) = \frac{1}{2\pi i} \oint_{\partial B_\delta(\mu_0)} (\mu - \hat{\chi}_s(z))^{-1}\, d\mu
$$
$$
= \frac{1}{2\pi i} \oint_{\partial B_\delta(\mu_0)} (\mu P_{V_j}^\perp - \hat{\chi}_{s,j}(z))^{-1}\, d\mu + \mathcal{O}(|z - \omega_j|),
$$

where $(\mu P_{V_j}^\perp - \hat{\chi}_{s,j}(z))^{-1}$ is the inverse with respect to $P_{V_j}^\perp$. In particular,

$$
\operatorname{rank} Q(z) = \dim \ker(\mu - \hat{\chi}_{s,j}(\omega_j)), \quad \text{for any } z \in U_\delta,
$$

and

$$
Q(z) P_{V_j} = \mathcal{O}(|z - \omega_j|) \quad \text{and} \quad P_{V_j} Q(z) = \mathcal{O}(|z - \omega_j|) \tag{46}
$$

for any $z \in U_\delta$.

Proof. The first step is to prove the following claim: for any $\mu_0 \in \mathbb{C}$ in the resolvent set of $\hat{\chi}_{s,j}(\omega_j)$ (where the resolvent/spectra is with respect to $\mathcal{B}(V_j^\perp)$), there exist neighborhoods U of ω_j and W of μ_0 such that $W \cap \sigma(\hat{\chi}_s(z)) = \emptyset$ for any $z \in U$.

So let $\mu_0 \in \mathbb{C}$ belong to the resolvent set of $\hat{\chi}_{s,j}(\omega_j)$. Then, from standard perturbation theory (see Lemma C.1) and the continuity of $z \mapsto \hat{\chi}_{s,j}(z)$, we can find $\delta > 0$ small enough such that $B_\delta(\mu_0) \subset \mathbb{C}$ lies on the resolvent set of $\hat{\chi}_{s,j}(z)$ for any z close enough to ω_j. In particular, the inverse $(\mu P_{V_j}^\perp - \hat{\chi}_{s,j}(z))^{-1}$ exists and is uniformly bounded for z close enough to ω_j and $\mu \in W := B_{\delta/2}(\mu_0)$. Therefore, we can apply Lemma 4.8 to $\mu - \hat{\chi}_s(z) = A(z, \mu) + (z - \omega_j)^{-1} B$, where

$$
A(z, \mu) = \mu - \hat{\chi}_s(z) + (z - \omega_j)^{-1} B_j, \quad B = B_j,
$$

and

$$
P_{V_j}^\perp A(z, \mu) P_{V_j}^\perp = \mu P_{V_j}^\perp - \hat{\chi}_{s,j}(z),
$$

to conclude that $\mu - \hat{\chi}_s(z)$ is invertible and

$$
(\mu - \hat{\chi}_s(z))^{-1} = (\mu P_{V_j}^\perp - \hat{\chi}_{s,j}(z))^{-1} + \mathcal{O}(|z - \omega_j|), \quad \text{for any } \mu \in W \text{ and } z \in U, \tag{47}
$$

where $(\mu P_{V_j}^\perp - \hat{\chi}_{s,j})^{-1}$ is the inverse with respect to $P_{V_j}^\perp$.

Next, let $0 < \mu_0 \in \sigma(\hat{\chi}_{s,j}(\omega_j))$. So, by the same arguments as in the proof of Proposition 4.4, we find that μ_0 belongs to the discrete spectrum of $\hat{\chi}_{s,j}(\omega_j)$. Thus, from classical perturbation theory again, we can find an annulus

$$
\overline{B_\delta(\mu_0) \setminus B_{\delta/2}(\mu_0)} \subset \{z \in \mathbb{C} : \operatorname{Re}(z) > 0\}
$$

contained in the resolvent set of $\hat{\chi}_{s,j}(z)$ for any z sufficiently close to ω_j. In particular, from our first claim and a compactness argument, we can take a small enough neighborhood of $z = \omega_j$ such that the annulus $\overline{B_\delta(\mu_0) \setminus B_{\delta/2}(\mu_0)}$ also lies inside the resolvent set of $\hat{\chi}_s(z)$. In this neighborhood, the spectral (Riesz) projection defined by

$$Q(z) = \frac{1}{2\pi i} \oint_{\partial B_\delta(\mu_0)} (\mu - \hat{\chi}_s(z))^{-1} \, d\mu$$

is holomorphic and has constant rank. Moreover, substituting $(\mu - \hat{\chi}_s(z))^{-1}$ by (47) in the above we obtain

$$Q(z) = \frac{1}{2\pi i} \oint_{\partial B_\delta(\mu_0)} (\mu P_{V_j}^\perp - \hat{\chi}_{s,j}(z))^{-1} \, d\mu + \mathcal{O}(|z - \omega_j|).$$

Moreover, since $(\mu P_{V_j}^\perp - \hat{\chi}_{s,j}(z))^{-1} P_{V_j} = P_{V_j}(\mu P_{V_j}^\perp - \hat{\chi}_{s,j}(z))^{-1} = 0$ we obtain (46).

To complete the proof we just need to show that rank $Q(z) = \dim \ker \mu_0 - \hat{\chi}_{s,j}(\omega_j)$. This follows from the fact that $Q(z)$ is a continuous family of projections, hence rank $Q(z)$ is constant, and $Q(z) \to \frac{1}{2\pi i} \oint (\mu P_{V_j}^\perp - \hat{\chi}_{s,j}(\omega_j))^{-1}$, which is the orthogonal projection on $\ker \mu_0 - \hat{\chi}_{s,j}(\omega_j)$ (as $\hat{\chi}_{s,j}(\omega_j)$ is symmetric).   ∎

We are now in a position to prove Proposition 4.2.

Proof of Proposition 4.2. Note that the set $\{z \in \mathcal{D}_\Omega \setminus \mathcal{P}(\hat{\chi}_s) : 1 \notin \sigma(\hat{\chi}(z))\} \subset \mathbb{C}$ is open by continuity. Hence, $(1 - \hat{\chi}_s(z))^{-1}$ is well defined and holomorphic on this set, and we just need to worry about the points where $1 - \hat{\chi}_s$ is not invertible, and the points ω_j where χ_s blows up. By Lemma 4.6, the set of points where $1 - \hat{\chi}_s$ is not invertible is contained in the intervals $\omega \in (-\Omega, -\omega_1] \cap [\omega_1, \Omega)$. Moreover, by the symmetries of $\hat{\chi}_s$, it is enough to look on the positive interval $[\omega_1, \Omega)$.

So first, let us consider the points $\omega \in (\omega_j, \omega_{j+1})$ for some $j \leq m$, where $1 - \hat{\chi}_s(\omega)$ is not invertible. For these points, we know from Proposition 4.4 that 1 belongs to the discrete spectrum of $\hat{\chi}_s(\omega)$. Hence, from Lemma 4.7, estimate (40), and the fact that $\hat{\chi}_s(\omega)$ is self-adjoint, we conclude that this set of points is discrete, and that $(1 - \hat{\chi}_s(\omega))^{-1}$ has a pole with rank equal to $\dim \ker(1 - \hat{\chi}_s(\omega))$ at any such point.

Next we want to show that any excitation ω_j is a pole of $(1 - \hat{\chi}_s)^{-1}$ with rank equal to $\dim \ker(1 - P_{V_j}^\perp \hat{\chi}_s(\omega_j) P_{V_j}^\perp)$. So let ω_j be a pole of $\hat{\chi}_s$, and let z be in a neighborhood of ω_j such that the projection

$$Q(z) = \frac{1}{2\pi i} \oint_{\partial B_\delta(1)} (\xi - \hat{\chi}_s(z))^{-1} \, d\xi$$

has rank $M_j = \dim \ker(1 - P_{V_j}^\perp \hat{\chi}_s(\omega_j) P_{V_j}^\perp)$. That this projection is well defined and holomorphic for z close to ω_j is a consequence of Lemma 4.9. Thus, since $\hat{\chi}_s(z)$ commutes with $Q(z)$, we can deal with the operators

$$1 - \hat{\chi}_s(z) = \underbrace{Q(z)(1 - \hat{\chi}_s(z))Q(z)}_{:=K(z)} + \underbrace{\tilde{Q}(z)(1 - \hat{\chi}_s(z))\tilde{Q}(z)}_{:=\tilde{K}(z)}$$

separately.

Let us start with $K(z)$. From the definition of $Q(z)$ and Lemma 4.9, we see that $K(z) \to 0$ as $z \to \omega_j$. Hence, $K(z)$ can be expanded as

$$K(z) = (z - \omega_j)K_1 + \mathcal{O}(|z - \omega_j|),$$

for some $K_1 \in \mathcal{B}(\mathcal{H})$. Moreover, from the blow-up estimate (40), we also have

$$\|K(\omega_j + i\eta)f\| \gtrsim |\eta|\|f\|, \quad \text{for any } f \in \operatorname{ran} Q(z) \text{ and } \eta \text{ small.}$$

Hence, the same arguments as the proof of Lemma 4.7 lead to the conclusion that K_1 is invertible with respect to $Q(\omega_j)$, that $K(z)$ is invertible with respect to $Q(z)$ for $z \neq \omega_j$, and that

$$K(z)^{-1} = (z - \omega_j)^{-1}K_{-1} + \mathcal{O}(1), \tag{48}$$

for some operator K_{-1} with rank $K_{-1} = M = \dim \ker(1 - P_{V_j}^{\perp}\hat{\chi}_s(\omega_j)P_{V_j}^{\perp})$. The big-O term here is with respect to the limit $z \to \omega_j$.

For $\widetilde{K}(z)$ we can use formula (66) in the appendix. Indeed, introducing the Riesz projection $Q_j(z) = \oint_{\partial B_\delta(\mu_0)}(\mu P_{V_j}^{\perp} - \hat{\chi}_{s,j}(z))^{-1}\, d\mu$ of $\hat{\chi}_{s,j}(z)$ around 1, then from the definition of $\widetilde{Q}(z)$, formula (66), and Lemma 4.8, we find that

$$\begin{aligned}
\widetilde{K}(z)^{-1} &= \frac{1}{2\pi i}\oint_{\partial B_\delta(1)} \frac{1}{\mu - 1}(\mu - \hat{\chi}_s(z))^{-1}\, d\mu \\
&= \frac{1}{2\pi i}\oint_{\partial B_\delta(1)} \frac{1}{\mu - 1}(\mu P_{V_j}^{\perp} - \hat{\chi}_{s,j}(z))^{-1}\, d\mu + \mathcal{O}(|z - \omega_j|) \\
&= \left((P_{V_j}^{\perp} - Q_j(z))(1 - \hat{\chi}_{s,j}(z))(P_{V_j}^{\perp} - Q_j(z))\right)^{-1} + \mathcal{O}(|z - \omega_j|) \\
&= \mathcal{O}(1).
\end{aligned} \tag{49}$$

Combining (48) and (49), we have shown that

$$(1 - \hat{\chi}_s(z))^{-1} = K^{-1} + \widetilde{K}^{-1} = \frac{K_{-1}}{z - \omega_j} + K_0(z),$$

where K_{-1} is invertible with respect to the orthogonal projection on $\ker 1 - \hat{\chi}_{s,j}(\omega_j)$ and the operator $K_0(z)$ is holomorphic and uniformly bounded around $z = \omega_j$.

To complete the proof, it is enough to show that $\operatorname{ran} K_0(\omega_j) \subset \operatorname{ran} K_{-1}$. This follows from the identity

$$\begin{aligned}
K_0(\omega_j)P_{V_j} &= \lim_{z \to \omega_j}(1 - \hat{\chi}_s(z))^{-1}P_{V_j} = \lim_{z \to \omega_j} K(z)^{-1}P_{V_j} + \underbrace{\lim_{z \to \omega_j}(\widetilde{K}(z))^{-1}P_{V_j}}_{=0} \\
&= \lim_{z \to \omega_j} Q(z)K(z)^{-1}P_{V_j} = Q(\omega_j)\lim_{z \to \omega_j} K(z)^{-1}P_{V_j},
\end{aligned}$$

where we used that $K_{-1}P_{V_j} = 0$, and $\widetilde{K}(z)^{-1}P_{V_j} = \mathcal{O}(|z - \omega_j|)$ (see (49)), and the fact that $\operatorname{ran} Q(\omega_j) = \operatorname{ran} K_{-1}$. ∎

4.2. Proof of Proposition 4.3

The strategy here is to show that the eigenvalues of $\hat{\chi}_s(\omega)$, as a function of ω, are strictly decreasing along the intervals (ω_j, ω_{j+1}), and then analyze what happens when ω crosses ω_j. For this, let us introduce the number of eigenvalues, counting multiplicity, greater than μ_0 as

$$n_{\mu_0}(\omega) := \sum_{\mu > \mu_0} \dim \ker(\mu - \hat{\chi}_s(\omega)).$$

Then the following lemma holds:

Lemma 4.10 (Strictly decreasing eigenvalues). *The positive eigenvalues of $\hat{\chi}_s(\omega)$ are decreasing functions of ω in the interval (ω_j, ω_{j+1}), $j \geq 0$. Moreover, for any $\mu_0 > 0$, we have*

$$n_{\mu_0}(\omega_j^+) = n_{\mu_0}(\omega_j^-) + \dim V_j - \dim \ker(\mu_0 - P_{V_j}^{\perp} \hat{\chi}_s(\omega_j) P_{V_j}^{\perp}), \qquad (50)$$

where $n_{\mu_0}(\omega_j^+) = \lim_{\omega \to \omega_j, \omega > \omega_j} n_{\mu_0}(\omega)$ and $n_{\mu_0}(\omega_j^-) = \lim_{\omega \to \omega_j, \omega < \omega_j} n_{\mu_0}(\omega)$ are, respectively, the right and left limits of $n_{\mu_0}(\omega)$ at ω_j.

Proof. We first note that since the function $h(\lambda, \omega) = \frac{2\lambda}{\omega^2 - \lambda^2}$ is decreasing for ω in the intervals $(-\infty, \lambda)$ and (λ, ∞), and since the spectral measure $\|P_{\lambda + E_0} T^* f\|^2$ is not identically zero for any f in the complement of the kernel of T^*, then for any $\omega > \omega'$ in (ω_j, ω_{j+1}),

$$\langle f, \hat{\chi}_s(\omega') f \rangle = \int_{\omega_1}^{\infty} h(\lambda, \omega') \, \mathrm{d} \| P_{\lambda + E_0}^{H} T^* f \|^2$$
$$> \int_{\omega_1}^{\infty} h(\lambda, \omega) \, \mathrm{d} \| P_{\lambda + E_0}^{H} T^* f \|^2 = \langle f, \hat{\chi}_s(\omega) f \rangle.$$

Hence by the Rayleigh–Ritz principle, the positive eigenvalues of $\hat{\chi}_s(\omega)$ are decreasing functions of ω in the interval (ω_j, ω_{j+1}).

The existence of the right and left limits of $n_{\mu_0}(\omega)$ at ω_j follows since $n_{\mu_0}(\omega)$ is decreasing. By Lemma 4.9 and the decreasing property of the eigenvalues of $\hat{\chi}_s$, the left limit of $n_{\mu_0}(\omega_j^-)$, $\omega < \omega_j$ is exactly the number of eigenvalues of $P_{V_j}^{\perp} \hat{\chi}_s(\omega_j) P_{V_j}^{\perp}$ equal to or greater than μ_0. Indeed, by Lemma 4.9, an eigenvalue of $P_{V_j}^{\perp} \hat{\chi}_s(\omega_j) P_{V_j}^{\perp}$ equal to or greater than μ_0 corresponds to another eigenvalue of $\hat{\chi}_s(\omega)$, for ω in a neighborhood of ω_j. Since $\omega < \omega_j$, by the decreasing property, the corresponding eigenvalue is greater than μ_0. Conversely, if $\mu(\omega)$ is an eigenvalue of $\hat{\chi}_s(\omega)$ such that $\lim_{\omega \to \omega_j, \omega < \omega_j} \mu(\omega) \geq \mu_0$, then as $\lim_{\omega \to \omega_j, \omega < \omega_j} \frac{2\omega_j}{\omega^2 - \omega_j^2} = -\infty$ the corresponding family of eigenfunctions has a vanishing component on V_j. Hence $\mu(\omega)$ converges to an eigenvalue of $P_{V_j}^{\perp} \hat{\chi}_s(\omega_j) P_{V_j}^{\perp}$.

For the right limit $n_{\mu_0}(\omega_j^+)$, consider an eigenvalue $\mu(\omega)$ of $\hat{\chi}_s(\omega)$, $\omega > \omega_j$ greater than μ_0. Since $\omega \mapsto \mu(\omega)$ is decreasing, it either diverges at ω_j or it converges to an eigenvalue of $P_{V_j}^{\perp} \hat{\chi}_s(\omega_j) P_{V_j}^{\perp}$ strictly greater than μ_0 by a similar argument to above. Finally,

there are exactly $\dim V_j$ eigenvalues blowing up at ω_j since for any $f \in V_j$, we have

$$\langle f, \hat{\chi}_s(\omega) f \rangle \geq \left\langle f, \frac{2\omega_j}{\omega^2 - \omega_j^2} B_j f \right\rangle - \left\| \hat{\chi}_s(\omega) - \frac{2\omega_j}{\omega^2 - \omega_j^2} B_j \right\| \|f\| \gtrsim \frac{2\omega_j}{\omega^2 - \omega_j^2} \|f\|^2,$$

so $\lim_{\omega \to \omega_j, \omega > \omega_j} \langle f, \hat{\chi}_s(\omega) f \rangle = \infty$ and $P_{V_j}^\perp \hat{\chi}_s(\omega) P_{V_j}^\perp$ is bounded in a neighborhood of ω_j.   ∎

Proof of Proposition 4.3. From Proposition 4.2, $1 - \hat{\chi}_s(\omega)$ is invertible for $0 < \omega < \omega_1$. Since $P_{V_1}^\perp \hat{\chi}_s(\omega) P_{V_1}^\perp$ is negative for $|\omega| < \omega_2$, from Lemma 4.9, ω_1 is not a pole of $(1 - \hat{\chi}_s)^{-1}$. By Proposition 2.9, we have $\operatorname{rank}_{\omega_j}(\hat{\chi}_s) = \dim V_j$, so by Lemma 4.6 it is enough to show that

$$\sum_{\omega_1 < \omega < \omega_{j+1}} \operatorname{rank}_\omega((1 - \hat{\chi}_s)^{-1}) \leq \sum_{k \leq j} \dim V_k \quad \text{for any } j \leq m. \tag{51}$$

By Proposition 4.2, $\operatorname{rank}_\omega((1 - \hat{\chi}_s)^{-1}) = \dim \ker(1 - \hat{\chi}_s(\omega))$ for any $\omega \in (\omega_j, \omega_{j+1})$. From the decreasing property of the eigenvalues in Lemma 4.10, the sum of the ranks of the poles in the interval (ω_j, ω_{j+1}) is given by the number of eigenvalues of $\hat{\chi}_s(\omega)$ that cross 1. Hence for any $j \geq 0$,

$$\sum_{\omega_j < \omega < \omega_{j+1}} \operatorname{rank}_\omega((1 - \hat{\chi}_s)^{-1}) = n_1(\omega_j^+) - n_1(\omega_{j+1}^-).$$

As a result, combining Lemma 4.9, estimate (50), and the rank characterization in (33) we get

$$\sum_{\omega_1 < \omega < \omega_{j+1}} \operatorname{rank}_\omega((1 - \hat{\chi}_s)^{-1})$$

$$= \sum_{k=1}^{j} \left(\operatorname{rank}_{\omega_k}((1 - \hat{\chi}_s)^{-1}) + \sum_{\omega_k < \omega < \omega_{k+1}} \operatorname{rank}_\omega((1 - \hat{\chi}_s)^{-1}) \right)$$

$$= \sum_{k=1}^{j} \left(\dim \ker(\mu_0 - P_{V_j}^\perp \hat{\chi}_s(\omega_j) P_{V_j}^\perp) + n_1(\omega_k^+) - n_1(\omega_{k+1}^-) \right)$$

$$= \sum_{k=1}^{j} n_1(\omega_k^-) + \dim V_k - n_1(\omega_{k+1}^-)$$

$$= n_1(\omega_1^-) - n_1(\omega_{j+1}^-) + \sum_{k=1}^{j} \dim V_k.$$

But since $n_1(\omega_{j+1}^+) \geq 0$ and $n_1(\omega_1^-) = 0$ as $\hat{\chi}_s(\omega)$ is nonpositive semidefinite for $\omega < \omega_1$, we obtain (51).   ∎

5. The Fourier transform of χ^{RPA}

We can now use Propositions 4.2 and 4.3 in the particular case $T = F_H^{1/2} S$ to prove Theorems 1.2 and 1.4.

Proof of Theorem 1.2. Let χ_0 be the density–density response function of some Hamiltonian satisfying Assumption 1 and let $\chi^{\mathrm{RPA}} = \mathcal{S}^{\mathrm{RPA}}(\chi_0)$ be the associated solution to the RPA-Dyson equation. Then, since $\|\chi_0(t)\|_{L^2+L^\infty, L^1\cap L^2}$ is uniformly bounded by Proposition 2.4, from the Grönwall inequality we know that $\|\chi^{\mathrm{RPA}}(t)\|_{L^2+L^\infty, L^1\cap L^2} \lesssim e^{Dt}$ for some $D > 0$. Hence, the Fourier transform $\widehat{\chi^{\mathrm{RPA}}}(z)$ is well defined for $\mathrm{Im}(z) > D$ and we have (by the convolution property of the Fourier transform)

$$\widehat{\chi^{\mathrm{RPA}}}(z) = \hat{\chi}_0(z) + \hat{\chi}_0(z) F_H \widehat{\chi^{\mathrm{RPA}}}(z), \quad \text{for } \mathrm{Im}(z) > D. \tag{52}$$

Let $\tilde{\chi}$, $\tilde{\chi}_0$, and $\hat{\chi}_s$ be the operators defined by

$$\tilde{\chi} := F_H^{1/2} \widehat{\chi^{\mathrm{RPA}}}, \quad \tilde{\chi}_0 := F_H^{1/2} \hat{\chi}_0, \quad \text{and} \quad \hat{\chi}_s := F_H^{1/2} \hat{\chi}_0 F_H^{1/2}, \tag{53}$$

where $F_H^{1/2} = \sqrt{4\pi}(-\Delta)^{-\frac{1}{2}}$ is up to a multiplicative constant the convolution against $\frac{1}{|\cdot|^2}$. Then we have

$$\tilde{\chi}(z) = \tilde{\chi}_0(z) + \hat{\chi}_s(z)\tilde{\chi}(z) \implies (1 - \hat{\chi}_s(z))\tilde{\chi}(z) = \tilde{\chi}_0(z).$$

Also, we see from the Hardy–Littlewood–Sobolev inequality that $\tilde{\chi}_0(z) \in \mathcal{B}(L^2(\mathbb{R}^3) + L^\infty(\mathbb{R}^3), L^2(\mathbb{R}^3))$, and that $\hat{\chi}_s$ is an operator of the form of equation (31) with $T = F_H^{1/2} S \in \mathcal{B}(\mathcal{H}_N, L^2(\mathbb{R}^3))$. Therefore, from Proposition 4.2, the map

$$z \mapsto (1 - \hat{\chi}_s(z))^{-1}\tilde{\chi}_0(z) \in \mathcal{B}(L^1(\mathbb{R}^3) \cap L^2(\mathbb{R}^3), L^2(\mathbb{R}^3))$$

is the unique meromorphic extension of $\tilde{\chi}$ to the domain $\mathcal{D}_\Omega = \{z \in \mathbb{C}: \mathrm{Im}(z) \neq 0 \text{ or } |\mathrm{Re}(z)| < \Omega\}$. Now going back to equation (52), we see from (53) that

$$\begin{aligned} \widehat{\chi^{\mathrm{RPA}}}(z) &= \hat{\chi}_0(z) + \hat{\chi}_0(z) F_H^{1/2} \tilde{\chi}(z) \\ &= \hat{\chi}_0(z) + \hat{\chi}_0(z) F_H^{1/2}(1 - \hat{\chi}_s(z))^{-1} F_H^{1/2} \hat{\chi}_0(z). \end{aligned} \tag{54}$$

In particular, $\widehat{\chi^{\mathrm{RPA}}}$ has a unique meromorphic extension as a map from $\mathcal{D}_\Omega$ to $\mathcal{B}(L^2 + L^\infty, L^1 \cap L^2)$, which proves item (i) from Theorem 1.2.

For the other item in Theorem 1.2, we want to relate the poles of $\widehat{\chi^{\mathrm{RPA}}}$ to the poles of $(1 - \hat{\chi}_s)^{-1}$. This can be done by observing that, since $F_H^{1/2}: L^1(\mathbb{R}^3) \cap L^2(\mathbb{R}^3) \to L^2(\mathbb{R}^3)$ is injective and bounded, a point $z \in \mathcal{D}_\Omega$ is a pole of $\widehat{\chi^{\mathrm{RPA}}}(z)$ with finite rank if and only if it is a pole of $\tilde{\chi} = F_H^{1/2} \widehat{\chi^{\mathrm{RPA}}}$ with the same rank. Therefore, if we can show that any pole of $\tilde{\chi}$ is simple and the inequality

$$\mathrm{rank}_\omega(\tilde{\chi}) \le \mathrm{rank}_\omega((1 - \hat{\chi}_s)^{-1}) \tag{55}$$

holds for any $0 < \omega < \Omega$ (with the convention that the rank is 0 if ω is not a pole), then Theorem 1.2 follows from the forward shift property in Proposition 4.3.

For $\omega \in \mathcal{P}((1 - \hat{\chi}_s)^{-1}) \setminus \mathcal{P}(\hat{\chi}_0)$, since $\hat{\chi}_0$ is holomorphic in a neighborhood of ω, it is clear from (54) that ω is at most a simple pole of $\widehat{\chi^{\mathrm{RPA}}}$ and that inequality (55) holds. We are thus left with the points in $\mathcal{P}(\hat{\chi}_0)$. Let $\omega_j \in \mathcal{P}(\hat{\chi}_0)$. Since $F_H^{1/2}$ is injective and bounded from $L^1(\mathbb{R}^3) \cap L^2(\mathbb{R}^3)$ to $L^2(\mathbb{R}^3)$, we have $\dim \operatorname{ran} F_H^{1/2} SP_{E_0+\omega_j}^H = \dim \operatorname{ran} SP_{E_0+\omega_j}^H$. This implies that the poles of $\hat{\chi}_s$ are precisely the poles of $\hat{\chi}_0$ and they have the same rank. Hence, using the spectral decomposition of H, we have that $\tilde{\chi}_0(z) + 2\frac{\omega_j}{\omega_j^2 - z^2} F_H^{1/2} SP_{E_0+\omega_j}^H S^*$ is bounded. By the decomposition (34) of $(1 - \hat{\chi}_s(z))^{-1}$ for z in a neighborhood of ω_j, we obtain

$$(z - \omega_j)\tilde{\chi}(z)$$
$$= (K_{-1} + (z - \omega_j)K_0)\left(\frac{F_H^{1/2} SP_{E_0+\omega_j}^H S^*}{z - \omega_j} + \tilde{\chi}_0 - \frac{F_H^{1/2} SP_{E_0+\omega_j}^H S^*}{z - \omega_j} \right)$$
$$\quad + \mathcal{O}(|z - \omega_j|)$$
$$= K_{-1}\frac{F_H^{1/2} SP_{E_0+\omega_j}^H S^*}{z - \omega_j} + K_{-1}\left(\tilde{\chi}_0 - \frac{F_H^{1/2} SP_{E_0+\omega_j}^H S^*}{z - \omega_j} \right) + K_0 F_H^{1/2} SP_{E_0+\omega_j}^H S^*$$
$$\quad + \mathcal{O}(|z - \omega_j|).$$

Recall that $\hat{\chi}_s$ is given by equation (31) by taking $T = F_H^{1/2}S$. Hence, using the last statement in Proposition 4.2, we get that $K_{-1} F_H^{1/2} SP_{E_0+\omega_j}^H S^* = 0$. This shows that ω_j is at most a simple pole of $\tilde{\chi}$. Moreover, using the last statement of Proposition 4.2 again, $\operatorname{ran} K_0 F_H^{1/2} SP_{E_0+\omega_j}^H \subset \operatorname{ran} K_{-1}$. As, by equation (33),

$$\dim \operatorname{ran} K_{-1} = \dim \ker(1 - P_{V_j}^{\perp}\hat{\chi}_s(\omega_j)P_{V_j}^{\perp}) = \operatorname{rank}_{\omega_j}((1 - \hat{\chi}_s)^{-1}),$$

the inequality $\operatorname{rank}_{\omega_j}(\tilde{\chi}) \leq \operatorname{rank}_{\omega_j}((1 - \hat{\chi}_s)^{-1})$ holds for any $j \leq m$ and the proof is complete. ∎

Notice that we have proved that $\operatorname{rank}_{\omega}(\widehat{\chi^{\mathrm{RPA}}}) \leq \operatorname{rank}_{\omega}((1 - \hat{\chi}_s)^{-1})$ for any $0 < \omega < \Omega$, which will be useful in the proof of Theorem 1.4.

Proof of Theorem 1.4. By the rank characterization in equation (33) from Proposition 4.2, it is enough to show that $\operatorname{rank}_{\omega}(\widehat{\chi^{\mathrm{RPA}}}) = \operatorname{rank}_{\omega}((1 - \hat{\chi}_s)^{-1})$ for any $0 < \omega < \Omega$. The inequality $\operatorname{rank}_{\omega}(\widehat{\chi^{\mathrm{RPA}}}) = \operatorname{rank}_{\omega}(\tilde{\chi}) \leq \operatorname{rank}_{\omega}((1 - \hat{\chi}_s))$ has already been proved . For the converse inequality, observe that, since $\tilde{\chi}: \mathcal{D}_{\Omega} \to \mathcal{B}(L^2 + L^{\infty}, L^2)$ is meromorphic, the map $\tilde{\chi}F_H^{1/2}: \mathcal{D}_{\Omega} \to \mathcal{B}(L^2, L^2)$ is also a meromorphic family of operators. Thus the inequality $\operatorname{rank}_{\omega}(\tilde{\chi}F_H^{1/2}) \leq \operatorname{rank}_{\omega}(\tilde{\chi})$ holds since the composition of linear operators can only lower the rank. The proof is now complete because

$$\tilde{\chi}(z)F_H^{1/2} = (1 - \hat{\chi}_s(z))^{-1}\underbrace{F_H^{1/2}\hat{\chi}_0(z)F_H^{1/2}}_{=\hat{\chi}_s(z)} = (1 - \hat{\chi}_s(z))^{-1} - 1, \tag{56}$$

which implies that $\operatorname{rank}_{\omega}((1 - \hat{\chi}_s)^{-1}) = \operatorname{rank}_{\omega}(\tilde{\chi}F_H^{1/2}) \leq \operatorname{rank}_{\omega}(\widehat{\chi^{\mathrm{RPA}}})$. ∎

Remark 5.1. Ideally, we would like to conclude the proof of Theorem 1.2 directly from identity (56). However, this is not possible because $F_H^{1/2} : L^2 \to L^2 + L^\infty$ is not surjective (nor is the image dense in $L^2 + L^\infty$), and some poles could in principle vanish when composing $\widehat{\chi^{\mathrm{RPA}}}$ with $F_H^{1/2}$ on the right.

A. Time-dependent density functional theory

In TDDFT, one postulates the existence of an (exact) time-dependent exchange-correlation potential[1] v_{xc} such that the solution $\Phi(t)$ of the time-dependent Schrödinger equation

$$\begin{cases} i\,\partial_t \Phi(t) = H_{\mathrm{eff}}(t)\Phi(t) & \text{for } t > 0, \\ \Phi(0) = \Phi_0, \end{cases} \tag{57}$$

where the effective noninteracting Hamiltonian is given by

$$H_{\mathrm{eff}}(t) = -\frac{1}{2}\Delta + \sum_{j=1}^{N} v(r_j) + \epsilon f(t) v_{\mathscr{P}}(r_j) + \rho^{\Phi}(t) * \frac{1}{|\cdot|}(r_j)$$
$$+ v_{\mathrm{xc}}[\rho^{\Phi}; \Psi_0; \Phi_0](t, r_j), \tag{58}$$

reproduces the time-dependent electronic density ρ^{Ψ} of the solution $\Psi(t)$ of the interacting evolution (1). The potential v_{xc} at time t depends not just on the density ρ^{Ψ} at time t, but on its past history at all times $t' \in [0, t]$. The initial state of the noninteracting system Φ_0 can be chosen arbitrarily as long as [16, Chapter 4] it reproduces the initial density ρ^{Ψ_0} and the initial divergence of the current density

$$\nabla \cdot j^{\Psi_0}(r) = N\nabla \cdot \mathrm{Im} \int_{(\mathbb{R}^3)^{N-1}} \overline{\Psi_0(r, r_2, \ldots, r_N)} \nabla_r \Psi_0(r, r_2, \ldots, r_N)\,\mathrm{d}r_1 \ldots \mathrm{d}r_N.$$

Note however that the exact xc potential depends on this choice.

A.1. Formal derivation of the Dyson equation

In typical applications of linear response theory, the state Ψ_0 is the ground state of the static interacting Hamiltonian governing evolution (1). In particular, for $\varepsilon = 0$ the time-dependent density ρ^{Ψ} satisfies $\rho^{\Psi}(t) = \rho^{\Psi_0}$ for all times $t \geq 0$. Consequently, if we choose the ground state of the noninteracting system Φ_0 to be the exact Kohn–Sham ground state reproducing the density ρ^{Ψ_0} (assuming it exists), the time-dependent xc potential reduces to the exact xc potential of static DFT, i.e.

$$v_{\mathrm{xc}}[\rho^{\Psi}; \Psi_0; \Phi_0] = v_{\mathrm{xc}}^{\mathrm{static}}[\rho^{\Psi_0}],$$

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[1]For proofs of the existence, respectively uniqueness up to a time-dependent constant of such a potential see [19, 23], and for contributions to the open question of whether the required assumptions hold for systems with Coulomb interaction see [16, Section 4.4.2] and [9].

where Φ_0 is the ground state of the noninteracting Kohn–Sham Hamiltonian

$$H_0 = -\frac{1}{2}\Delta + \sum_{j=1}^{N} v(r_j) + \rho^{\Psi_0} * \frac{1}{|\cdot|}(r_j) + v_{\text{xc}}^{\text{static}}[\rho^{\Psi_0}](r_j). \tag{59}$$

From this observation we can now derive the Dyson equation of TDDFT. To this end, let us assume that the function $\rho \mapsto v_{\text{xc}}[\rho; \Phi_0; \Psi_0]$ is differentiable with respect to time-dependent densities at the stationary density $\rho(t) = \rho^{\Psi_0}$ for all t. Then, using the expansion of the electronic density given in equation (8), we have

$$v_{\text{xc}}[\rho^{\Psi}; \Psi_0; \Phi_0](r, t) = v_{\text{xc}}^{\text{static}}[\rho^{\Psi_0}](r) + \varepsilon F_{\text{xc}}(\chi \star f v_{\mathscr{P}})(r, t) + \mathcal{O}(\varepsilon^2),$$

where $F_{\text{xc}} = \frac{\delta v_{\text{xc}}}{\delta \rho}[\rho^{\Psi_0}; \Psi_0; \Phi_0]$ is a linear operator whose (Schwartz) kernel is called the (exact) exchange-correlation kernel of TDDFT. Similarly, the Hartree term can be expanded in powers of ε and the effective Hamiltonian (58) becomes

$$H_{\text{eff}}(t) = H_0 + \sum_{j=1}^{N} \varepsilon(F_H + F_{\text{xc}})(\chi \star f v_{\mathscr{P}})(r_j, t) + \varepsilon f(t) v_{\mathscr{P}}(r_j) + \mathcal{O}(\varepsilon^2),$$

where H_0 is the Kohn–Sham Hamiltonian (59) and F_H is the Hartree operator, acting on time-dependent potentials v by time-instantaneous convolution in space against the Coulomb potential,

$$(F_H v)(t, r) = \frac{1}{|\cdot|} * v(t, \cdot)(r) = \int_{\mathbb{R}^3} \frac{1}{|r - r'|} v(t, r') \, dr'.$$

The equivalence of the densities and (8) now yield

$$(\chi \star f v_{\mathscr{P}}) = \lim_{\varepsilon \to 0^+} \frac{\rho^{\Psi(t)} - \rho^{\Psi_0}}{\varepsilon} = \lim_{\varepsilon \to 0^+} \frac{\rho^{\Phi(t)} - \rho^{\Phi_0}}{\varepsilon}$$
$$= \chi_0 \star f v_{\mathscr{P}} + \chi_0 \star (F_H + F_{\text{xc}})(\chi \star f v_{\mathscr{P}}),$$

where χ and χ_0 are the density–density response functions of the interacting Hamiltonian and the noninteracting Kohn–Sham Hamiltonian, respectively. As the identity above holds for every time-dependent potential $f(t) v_{\mathscr{P}}(r)$, we obtain the celebrated Dyson equation of TDDFT,

$$\chi = \chi_0 + \chi_0 \star (F_H + F_{\text{xc}})\chi. \tag{60}$$

Remark A.1. Recalling that χ is a time-dependent operator on potentials, and making the convolution in time in equation (60) and the Hartree operator explicit, the equation says that for any time $t \geq 0$ and any potential $v_{\mathscr{P}} \in L^\infty(\mathbb{R}^3)$,

$$\chi(t) v_{\mathscr{P}} = \chi_0(t) v_{\mathscr{P}} + \int_0^t \chi_0(t - s) \frac{1}{|\cdot|} * (\chi(s) v_{\mathscr{P}}) + \chi_0(t - s) F_{\text{xc}}(\chi v_{\mathscr{P}})(s) \, ds,$$

where $\chi v_{\mathscr{P}}$ is the continuous $L^1(\mathbb{R}^3)$-valued map $s \mapsto \chi(s) v_{\mathscr{P}}$.

A.2. Common approximations

In the absence of any explicit description of the exact time-dependent xc potential v_{xc}, all practical TDDFT calculations must resort to approximations. The two most common ones are the following:

(1) *Random phase approximation (RPA).* Electron–electron interaction is only taken into account on a mean field level, that is to say, in (57)–(58) one only keeps the Hartree term but takes $v_{xc} = 0$. It follows that $F_{xc} = 0$ and the Dyson equation (60) reduces to the RPA-Dyson equation

$$\chi = \chi_0 + \chi_0 \star F_H \chi \tag{61}$$

treated in this paper.

(2) *Adiabatic local density approximation (ALDA).* In (57)–(58) one uses the instantaneous (or static) local density approximation at each time t,

$$v_{xc}[\rho^\Phi; \Psi_0; \Phi_0](r, t) = v_{xc}^{LDA}[\rho^{\Phi(t)}](r), \tag{62}$$

where v_{xc}^{LDA} is the LDA *exchange-correlation potential*, that is to say, $v_{xc}^{LDA}[\rho](r) = e_{xc}'(\rho(r))$, $e_{xc}(\bar\rho)$ is the exchange-correlation energy density per unit volume of a homogeneous electron gas with density $\bar\rho$ (known accurately from asymptotic and numerical results [5, 17]), and $e_{xc}'(\bar\rho) = \frac{d}{d\bar\rho} e_{xc}(\bar\rho)$. Since F_{xc} is the functional derivative of the exchange-correlation potential with respect to the density, it is given in this case by the multiplication operator

$$F_{xc}^{ALDA}(f v_\mathscr{P})(t, r) = \frac{\delta v_{xc}^{LDA}}{\delta \rho}[\rho^{\Phi_0}](f v_\mathscr{P})(r, t) = e''_{xc}(\rho^{\Phi_0}(r)) f(t) v_\mathscr{P}(r).$$

All the results in this paper (Theorems 1.1, 1.2, and 1.4) apply to equation (61). Note that the particular approximation chosen for the static xc potential v_{xc}^{static} underlying the reference response function χ_0 can be arbitrary, provided the Hamiltonian (59) satisfies Assumption 1.

In the approximation (62), the time-dependent xc potential only depends instantaneously on the electronic density $\rho(\cdot, t)$. This is an adiabatic approximation and it is one of the main challenging problems in TDDFT to improve on it in a systematic way [22, Chapter 8].

B. Poles of the density–density response function of a noninteracting Hamiltonian

Proposition B.1. *Let χ be the density–density response function defined in (21) of a noninteracting Hamiltonian H satisfying Assumption 1, i.e. $H = -\frac{1}{2}\Delta + \sum_{i=1}^{N} v(r_i)$. Let h be the one-body Hamiltonian $h = -\frac{1}{2}\Delta + v$ and $(\epsilon_k, \psi_k)_{1 \le k \le N} \in (\mathbb{R} \times L^2(\mathbb{R}^3))^N$*

be its lowest eigenpairs. Then we have for all $v_{\mathscr{P}} \in L^2(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ and $z \in \mathbb{C}$ with $\mathrm{Im}(z) > 0$,

$$\hat{\chi}(z)v_{\mathscr{P}}(r) = \sum_{k=1}^{N} \overline{\psi_k}(r)[(\epsilon_k - z - h)^{-1} + (\epsilon_k + z - h)^{-1}](v_{\mathscr{P}}\psi_k)(r). \tag{63}$$

Proof. The proof proceeds by deriving and exploiting explicit one-body expressions for the many-body operators appearing on the right-hand side of (24).

Step 1. We first show that

$$((E_0 - z - H)^{-1} + (E_0 + z - H)^{-1})S^* v_{\mathscr{P}}$$

$$= \frac{1}{\sqrt{N!}} \sum_{j=1}^{N} \sum_{\sigma \in S_N} (-1)^\sigma [(\epsilon_{\sigma(j)} - z - h)^{-1} + (\epsilon_{\sigma(j)} + z - h)^{-1}]v_{\mathscr{P}}(r_j)$$

$$\times \prod_{k=1}^{N} \psi_{\sigma(k)}(r_k). \tag{64}$$

Since $H\Psi_0 = E_0\Psi_0$, we have

$$((E_0 - z - H)^{-1} + (E_0 + z - H)^{-1})S^* v_{\mathscr{P}}$$

$$= ((E_0 - z - H)^{-1} + (E_0 + z - H)^{-1}) \sum_{i=1}^{N} v_{\mathscr{P}}(r_i)\Psi_0(r_1, \ldots, r_N).$$

Using that for any bounded continuous g, we have

$$\int_{\mathbb{R}} g(\lambda)\,\mathrm{d}P_\lambda^H = \int_{\mathbb{R}^N} g(\mu_1 + \cdots + \mu_N)\,\mathrm{d}P_{\mu_1}^h \otimes \cdots \otimes \mathrm{d}P_{\mu_N}^h,$$

and noticing that for $\sigma \in S_N$, $E_0 = \sum_{k=1}^{N} \epsilon_{\sigma(k)}$, we get (64).

Step 2. We have

$$S((E_0 - z - H)^{-1} + (E_0 + z - H)^{-1})S^* v_{\mathscr{P}}(r)$$

$$= \frac{N}{\sqrt{N!}} \int_{\mathbb{R}^{3(N-1)}} \overline{\Psi_0}(r, r_2, \ldots, r_N)$$

$$\times \sum_{\sigma \in S_N} (-1)^\sigma [(\epsilon_{\sigma(j)} - z - h)^{-1} + (\epsilon_{\sigma(j)} + z - h)^{-1}]$$

$$\times (v_{\mathscr{P}}(r)\psi_{\sigma(1)}(r)) \prod_{k \geq 2} \psi_{\sigma(k)}(r_k)\,\mathrm{d}r_2 \ldots \mathrm{d}r_N$$

$$- \left\langle \Psi_0, \frac{1}{\sqrt{N!}} \sum_{j=1}^{N} \sum_{\sigma \in S_N} (-1)^\sigma [(\epsilon_{\sigma(j)} - z - h)^{-1} + (\epsilon_{\sigma(j)} + z - h)^{-1}]v_{\mathscr{P}}(r_j) \right.$$

$$\left. \times \prod_{k=1}^{N} \psi_{\sigma(k)}(r_k) \right\rangle \rho_0(r).$$

Since $(h - \epsilon_{\sigma(j)})\psi_{\sigma(j)} = 0$, the second term on the right-hand side of the above equation vanishes. Thus, by orthonormality of $(\psi_i)_{1 \le i \le N}$, we obtain

$$\hat{\chi}(z)v_{\mathscr{P}}(r) = \sum_{k=1}^{N} \overline{\psi_k}(r)[(\epsilon_k - z - h)^{-1} + (\epsilon_k + z - h)^{-1}](v_{\mathscr{P}}\psi_k)(r). \qquad \blacksquare$$

We notice that expression (63) is equivalent to the Lehmann representation typically found in the physics and chemistry literature if, as is often assumed in this literature (but not strictly speaking valid for molecular Hamiltonians due to the presence of continuous spectrum), h is diagonalizable in an orthonormal basis $(\psi_i)_{i \in \mathbb{N}}$. Under this assumption,

$$[(\epsilon_k - z - h)^{-1} + (\epsilon_k + z - h)^{-1}](v_{\mathscr{P}}\psi_k)(r)$$
$$= \sum_{a=1}^{\infty} \Big(\frac{1}{\epsilon_k - \epsilon_a - z} + \frac{1}{\epsilon_k - \epsilon_a + z} \Big) \langle \psi_a, v_{\mathscr{P}}\psi_k \rangle \psi_a(r).$$

Hence, for $v_{\mathscr{O}} \in L^2(\mathbb{R}) + L^\infty(\mathbb{R}^3)$, we obtain

$$\langle v_{\mathscr{O}}, \hat{\chi}(z)v_{\mathscr{P}} \rangle$$
$$= \sum_{k=1}^{N} \sum_{a=1}^{\infty} \Big(\frac{1}{\epsilon_k - \epsilon_a - z} + \frac{1}{\epsilon_k - \epsilon_a + z} \Big) \langle \psi_k v_{\mathscr{O}}, \psi_a \rangle \langle \psi_a, v_{\mathscr{P}}\psi_k \rangle$$
$$= \sum_{k=1}^{N} \sum_{a=N+1}^{\infty} \Big(\frac{1}{\epsilon_k - \epsilon_a - z} + \frac{1}{\epsilon_k - \epsilon_a + z} \Big) \langle \psi_k v_{\mathscr{O}}, \psi_a \rangle \langle \psi_a, v_{\mathscr{P}}\psi_k \rangle. \qquad (65)$$

Note that the Lehmann representation (65) could also have been easily obtained from (27), by using the fact that the excited states of the noninteracting Hamiltonian H are given by the Slater determinants of the eigenstates of h containing at least one unoccupied eigenstate ψ_a, $a > N$, and that only those Slater determinants containing exactly one unoccupied eigenstate yield nonzero matrix elements in (27).

Equation (65) shows that the poles of the density–density response function of a non-interacting Hamiltonian are located at the spectral gaps between occupied and virtual eigenvalues of the one-body Hamiltonian.

C. Spectral theory of bounded operators

We collect here some results on the spectral theory of bounded operators on Banach spaces. These results are elementary and complete proofs can be found in [10]. For the convenience of the reader, we briefly sketch these proofs here.

Lemma C.1 (Continuity of spectra). *Let $A \in \mathscr{B}(E)$, where E is a Banach space and $\mu \in \rho(A)$ (where $\mathscr{B}(E)$ denotes the set of bounded linear operators on E and $\rho(A)$ denotes*

the resolvent set of A). Then for any B with $\|B\| < \|(\mu - A)^{-1}\|$ we have $\mu \in \rho(A + B)$. In particular, if $A\colon B_\delta(0) \subset \mathbb{C} \to \mathcal{B}(E)$ is continuous and $W \subset \rho(A(0))$ is compact, then $W \subset \rho(A(z))$ for any z close enough to 0.

Proof. For $\mu \in \rho(A)$, we have

$$(\mu - A)^{-1}(\mu - A - B) = I - (\mu - A)^{-1}B$$

and

$$(\mu - A - B)(\mu - A)^{-1} = I - B(\mu - A)^{-1}.$$

Thus, for $\|B\| < \|(\mu - A)^{-1}\|^{-1}$ the operators above are of the form $I - K$ with $\|K\| < 1$. Hence, they are invertible and the inverse is given by the Neumann series, $\sum_{k \in \mathbb{N}} K^n$.

For the second statement, note that since W is compact, and $(\mu - A(0))^{-1}$ is continuous with respect to μ, we have $\epsilon = \inf_{\mu \in W} \|(\mu - A(0))^{-1}\|^{-1} > 0$. Thus, from continuity we have $\|A(z) - A(0)\| \leq \epsilon$ for any z close to 0 and the result follows from the first statement. ∎

Lemma C.2 (Riesz projection and separation of spectra). *Let $\gamma \subset \rho(A) \subset \mathbb{C}$ be a closed smooth curve separating the spectrum of A. Then the operator*

$$P = \frac{1}{2\pi i} \oint_\gamma (\mu - A)^{-1} \mathrm{d}\mu$$

is a projection commuting with A. Moreover, for $\mu_0 \notin \gamma$, the operator

$$S(\mu_0) = \frac{1}{2\pi i} \oint_\gamma \frac{1}{\mu - \mu_0} (\mu - A)^{-1}(1 - P) \, \mathrm{d}\mu$$

satisfies

$$S(\mu_0) = \begin{cases} ((1 - P)(\mu_0 - A)(1 - P))^{-1} & \text{for } \mu_0 \text{ inside } \gamma, \\ (P(\mu_0 - A)P)^{-1} & \text{for } \mu_0 \text{ outside } \gamma. \end{cases} \tag{66}$$

In particular, the spectra of $A|_{\mathrm{ran}\,P} \in \mathcal{B}(\mathrm{ran}\,P)$ and $A|_{\ker P} \in \mathcal{B}(\ker P)$ are given respectively by the spectrum of A inside and outside γ.

Proof. That the operator P is well defined and bounded is clear since $\gamma \subset \rho(A)$ and $\mu \mapsto (\mu - A)^{-1}$ is continuous in μ. To see that P is a projection, note that one can choose a curve γ_1 inside γ such that all points lying between γ_1 and γ are in the resolvent of A. Thus, from a standard argument of holomorphic function theory,

$$P = \frac{1}{2\pi i} \oint_{\gamma_1} (\lambda - A)^{-1} \, \mathrm{d}\lambda.$$

Hence, multiplying the above by the definition of P with a contour integral on γ, using the resolvent identity $(\mu - A)^{-1}(\lambda - A)^{-1} = (\mu - \lambda)^{-1}((\lambda - A)^{-1} - (\mu - A)^{-1})$, and using the Cauchy integral formula for holomorphic functions, one can show that $P^2 = P$.

Since $S(\mu_0)$ commutes with A, formula (66) follows from

$$(\mu_0 - A)S(\mu_0) = \frac{1}{2\pi i} \oint_\gamma \frac{1}{\mu - \mu_0} - \frac{1}{2\pi i} \oint_\gamma (\mu - A)^{-1}\, \mathrm{d}\mu = \begin{cases} 1 - P & \text{for } \mu_0 \text{ inside } \gamma, \\ -P & \text{for } \mu_0 \text{ outside } \gamma, \end{cases}$$

and

$$S(\mu_0)P = \frac{1}{2\pi i} \oint_\gamma \oint_{\gamma_1} \frac{(\mu - A)^{-1} - (\lambda - A)^{-1}}{(\mu - \mu_0)(\lambda - \mu)}\, \mathrm{d}\lambda\, \mathrm{d}\mu = \begin{cases} 0 & \text{for } \mu_0 \text{ inside } \gamma, \\ S(\mu_0) & \text{for } \mu_0 \text{ outside } \gamma. \end{cases}$$

Finally, the last statement follows from two observations. First, the existence of the inverses in (66) implies that $\sigma(A|_{\mathrm{ran}\,P})$ lies inside γ and $\sigma(A|_{\ker P})$ lies outside γ. Second, from the decomposition

$$A = \begin{pmatrix} A|_{\mathrm{ran}\,P} & 0 \\ 0 & A|_{\ker P} \end{pmatrix}$$

with respect to $\mathcal{H} = \mathrm{ran}\,P \oplus \ker P$, we have $\sigma(A) = \sigma(A|_{\ker P}) \cup \sigma(A|_{\mathrm{ran}\,P})$. ∎

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Thiago Carvalho Corso
Department of Mathematics, Technische Universität München, Boltzmannstraße 3,
85748 Garching, Germany; thiago.carvalho-corso@ians.uni-stuttgart.de

Mi-Song Dupuy
Laboratoire Jacques-Louis Lions, Sorbonne Université, Boîte courrier 187, 75252 Paris, France;
mi-song.dupuy@sorbonne-universite.fr

Gero Friesecke
Department of Mathematics, Technische Universität München, Boltzmannstraße 3,
85748 Garching, Germany; gf@ma.tum.de