Projection operator approach to lifetimes of electrons in metals

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We present an alternative approach to the calculation of the lifetime of a single excited electron (hole) which interacts with the Fermi sea of electrons in a metal. The metal is modeled on the level of a Hamilton operator comprising a pertinent dispersion relation and scattering term. To determine the full relaxation dynamics we employ an adequate implementation of the time-convolutionless projection operator method. This yields an analytic expression for the decay rate which allows for an intuitive interpretation in terms of scattering events. It may furthermore be efficiently evaluated by means of a Monte Carlo integration scheme. As an example we investigate aluminum using, just for simplicity, a jellium-type model. This way we obtain data which are directly comparable to results from a self-energy formalism. Our approach applies to arbitrary temperatures.

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I. INTRODUCTION

For several decades, the dynamics of excited electrons in metals has been the subject of intense research in theoretical and experimental solid-state physics.[1–10] These investigations are motivated by the fact that a lot of physical and chemical properties of metallic materials depend essentially on those dynamics.[11–13] Lifetimes of (photo)excited electrons in metals are always short (on the order of femtoseconds) but the immense progress in ultrafast laser technology now allows for an experimental determination of such times, in which corresponding investigations are ongoing.[14]

Today a number of methods are used to calculate lifetimes of electrons. Practically all of them are formulated within the framework of Green's functions (many-body theory) and aim at determining the self-energy, particularly its imaginary part.[15–17] Many of them employ a screened interaction ("W") and a truncated expansion of the self-energy in terms of this screened interaction ("GW approximation").[7,8,18–27] The screened interaction is frequently obtained through a "random-phase approximation" (RPA).[11] For a simple sufficiently dense homogeneous gas of electrons interacting through Coulomb repulsion (jellium model), an approach along the above scheme is even feasible analytically and yields a closed expression for the lifetimes close to the Fermi edge (see below).[1,2] In a certain sense [which is described in more detail below, Eq. (17)], this approach leads to lifetimes which may quantitatively be compared to experimental data on, e.g., aluminum.[4,23,27] Of course timely state of the art approaches go beyond jellium and exploit not only the traditional self-energy formalism but also density-functional methods, etc.[23–26]

Our approach is, in contrary, not based on Green's functions at all but on projection operator techniques. A main motivation of our work is to demonstrate that a pertinent projective approach[28–35] is also capable in producing quantitative results on lifetimes. Our central formula from which the lifetimes are eventually calculated is in accord with expressions that may be derived within the above many-body approach [see below, Eq. (16)]. Furthermore it allows for an interpretation in terms of scattering events. This encourages further development of projection techniques as alternative quantitative tools for the investigation of relaxation and transport dynamics in condensed-matter systems (find more on this at the end of Sec. IV). However, our approach starts from an effective model comprising pertinent quasiparticle dispersion relations and an appropriate screened interaction. The (generically subtle) provision of such a suitable effective model is not part of our present analysis; the effective model thus has to be supplied by other means.

The paper at hand is organized as follows: in Sec. II we give a very brief introduction to the time-convolutionless projection operator method[35] and apply it to a general interacting quantum gas thus obtaining an expression for the electronic lifetime. In Sec. III we evaluate this expression numerically for a "screened" jellium model tuned to describe aluminum. We compare our results to other available data and comment on computing times. Eventually we close with discussion, summary, and outlook.

II. PROJECTIVE APPROACH TO OCCUPATION NUMBER DYNAMICS IN INTERACTING QUANTUM GASES

To determine the lifetime of an electron initially occupying some momentum eigenstate, we analyze the dynamics of the corresponding occupation number. A formalism which allows for such an analysis is the TCL method.[35] In general the latter is a perturbative projection operator technique which produces autonomous equations of motion for the variables of interest ("relevant information"). The technique may be applied to quantum system with a Hamiltonian of the type $\hat{H}=\hat{H}_0+\lambda\hat{V}$, where λ has to be in some sense small.[35] In order to apply this method one first has to construct a suitable projection operator $\mathcal{P}$. Formally, this is a linear map which projects any density matrix $\varrho(t)$ to a matrix $\mathcal{P}\varrho(t)$ that is determined by a certain set of variables. These variables should match with the variables of interest. Moreover $\mathcal{P}$ has to fulfill the property of a projection operator, that is, $\mathcal{P}^2=\mathcal{P}$. For initial states with $\mathcal{P}\varrho(0)=\varrho(0)$ the TCL scheme leads to a time-local differential equation for the dynamics of $\mathcal{P}\varrho$:

$$\partial_t \mathcal{P}\varrho(t) = \Gamma(t)\mathcal{P}\varrho(t), \quad \Gamma(t) = \sum_{k=1}^{\infty} \lambda^k \Gamma_k(t), \tag{1}$$

where the perturbative expansion used in the last equations is in principle exact. However, for a description to leading order, which is, typically and in our case, the second order, one has to determine $\Gamma_2(t)$. Whether or not a leading-order description will yield a reasonable result is a somewhat subtle question[36] but the expansion is well controlled and systematic, i.e., in principle higher order terms could be incorporated in a straightforward manner.[35] A widely accepted indicator for the validity of the truncation is a clear time scale separation between the resulting relaxation dynamics and the decay of the correlation function, the latter being introduced below. However, here we are going to focus on the leading order and comment on the time scales below when we eventually arrive at concrete lifetimes. In the literature[35] one finds

$$\Gamma_2(t) = \int_0^t dt' \mathcal{P}\mathcal{L}(t)\mathcal{L}(t')\mathcal{P}, \tag{2}$$

with $\mathcal{L}(t) = \frac{\imath}{\hbar}[\hat{V}(t), \#]$, where # denotes a placeholder for an operator which shall be inserted into the commutator. $V(t)$ refers to a perturbation in the interaction picture. With Eqs. (1) and (2) we obtain

$$\partial_t \mathcal{P}\varrho(t) = \int_0^t dt' \mathcal{P}\mathcal{L}(t)\mathcal{L}(t')\mathcal{P}\varrho(t). \tag{3}$$

Now for a concrete application we have to specify the underlying quantum model and a suitable projection operator.

The systems we investigate are interacting quantum gases, here of the "spinless fermions" type. The corresponding Hamiltonians may be written as

$$\underbrace{\sum_{\mathbf{k}} \varepsilon_{\mathbf{k}} a_{\mathbf{k}}^\dagger a_{\mathbf{k}}}_{\hat{H}_0} + \underbrace{\frac{1}{2}\sum_{\mathbf{k},\mathbf{l},\mathbf{q}} V(\mathbf{q}) a_{\mathbf{k}+\mathbf{q}}^\dagger a_{\mathbf{l}-\mathbf{q}}^\dagger a_{\mathbf{l}} a_{\mathbf{k}}}_{\hat{V}}, \tag{4}$$

where $\varepsilon_{\mathbf{k}}$ denotes a dispersion relation of free particles and $V(\mathbf{q})$ is the matrix elements of an interaction which depends on the concrete system. This Hamiltonian is of the above mentioned form as long as the interaction term $\hat{V}$ is in adequate sense "small" (see below). As will be demonstrated below (cf. Sec. III) it is thus reasonable to choose for $\varepsilon_{\mathbf{k}}$ pertinent quasiparticle dispersion relations and particularly for $V(\mathbf{q})$ an adequate screened interaction. Note that we neglect the spin quantum number since the system we consider in this work is paramagnetic, and without any magnetic fields the dispersion relation is the same for both spin channels. Below we are going to take care of this "spin degeneracy" in a very simple form [cf. text following Eq. (18)]. For the noninteracting many-particle system we may directly write down the wave-number (momentum) dependent "single-particle equilibrium density operator" as

$$\varrho_{\mathbf{j}}^{\mathrm{eq}} := f_{\mathbf{j}}(\mu,T) a_{\mathbf{j}}^\dagger a_{\mathbf{j}} + [1 - f_{\mathbf{j}}(\mu,T)] a_{\mathbf{j}} a_{\mathbf{j}}^\dagger, \tag{5}$$

with $f_{\mathbf{j}}(\mu,T) = \{\exp[(\varepsilon(\mathbf{j}) - \mu)/k_B T] + 1\}^{-1}$ being the Fermi distribution. Since we are interested in temperature regimes close to $T=0$ K but still $T \neq 0$ K, we can set the chemical potential $\mu \approx \varepsilon_F$. Further we abbreviate $f_{\mathbf{j}}(\varepsilon_F, T)$ as $f_{\mathbf{j}}$. The equilibrium density operator, again for the noninteracting case, of the total system, ϱ^{eq}, may be written as the tensor product of the single-particle density operators, i.e.,

$$\varrho^{\mathrm{eq}} := \bigotimes_{\mathbf{i}} \varrho_{\mathbf{i}}^{\mathrm{eq}}, \quad \text{note also} \quad \tilde{\varrho} := \bigotimes_{\mathbf{i}\neq\mathbf{j}} \varrho_{\mathbf{i}}^{\mathrm{eq}}. \tag{6}$$

Here, for later reference, $\tilde{\varrho}$ denotes the total density operator of the system which does not contain the subspace with respect to the momentum mode $\mathbf{j}$, i.e., it is $\varrho^{\mathrm{eq}} = \tilde{\varrho} \otimes \varrho_{\mathbf{j}}^{\mathrm{eq}}$. We should, also for later reference, mention here that while ϱ^{eq} is strictly speaking just the equilibrium state of the noninteracting system, it is routinely considered to describe the single-particle properties of the weakly interacting system more or less correctly. Thus, if single-particle observables relax toward equilibrium due to the interactions (scattering), we expect them to relax toward values corresponding to ϱ^{eq}.

For the investigations of the dynamics of excited states we define an operator $\Delta_{\mathbf{j}}$ (for the remainder of this paper $|\mathbf{j}\rangle$ denotes the excited state) as

$$\Delta_{\mathbf{j}} := (1 - f_{\mathbf{j}}) a_{\mathbf{j}}^\dagger a_{\mathbf{j}} - f_{\mathbf{j}} a_{\mathbf{j}} a_{\mathbf{j}}^\dagger = a_{\mathbf{j}}^\dagger a_{\mathbf{j}} - f_{\mathbf{j}}, \tag{7}$$

which describes the deviation of the mode occupation number $n_{\mathbf{j}} = a_{\mathbf{j}}^\dagger a_{\mathbf{j}}$ from its thermal equilibrium. Now, in order to apply the TCL method to this model we construct a suitable projector as follows:

$$\mathcal{P}\varrho(t) = \varrho^{\mathrm{eq}} + \frac{1}{\sigma_{\mathbf{j}}^2} \mathrm{Tr}\{\Delta_{\mathbf{j}} \varrho(t)\} \tilde{\varrho} \otimes \Delta_{\mathbf{j}}, \tag{8}$$

with $\varrho(t)$ being the density operator which describes the actual state of the system, $d_{\mathbf{j}}(t) := \mathrm{Tr}\{\Delta_{\mathbf{j}} \varrho(t)\}$ denotes the time-dependent expectation value of $\Delta_{\mathbf{j}}$, and $\sigma_{\mathbf{j}}^2 := (1-f_{\mathbf{j}})^2 + f_{\mathbf{j}}^2 = \mathrm{Tr}\{\Delta_{\mathbf{j}}^2\}$. It is straightforward to show that with the above definitions $\mathcal{P}$ is a projector and fulfills $\mathcal{P}^2\varrho(t) = \mathcal{P}\varrho(t)$. Note that $\mathrm{Tr}\{\varrho^{\mathrm{eq}} \Delta_{\mathbf{j}}\} = 0$ and $\mathrm{Tr}\{\Delta_{\mathbf{j}} \tilde{\varrho} \otimes \Delta_{\mathbf{j}}\} = \sigma_{\mathbf{j}}^2$. Before we eventually concretely apply Eq. (3) to our model we make the following approximation for an expression that appears in the computation of Eq. (3):

$$\begin{aligned} \mathcal{L}(t')\mathcal{P}\varrho(t) &= \frac{\imath}{\hbar}[\hat{V}(t'), \varrho^{\mathrm{eq}}] + \frac{\imath}{\hbar}[\hat{V}(t'), \tilde{\varrho} \otimes \Delta_{\mathbf{j}}] \frac{d_{\mathbf{j}}(t)}{\sigma_{\mathbf{j}}^2} \\ &\approx \frac{\imath}{\hbar}[\hat{V}(t'), \tilde{\varrho} \otimes \Delta_{\mathbf{j}}] \frac{d_{\mathbf{j}}(t)}{\sigma_{\mathbf{j}}^2}, \end{aligned} \tag{9}$$

The neglected commutator term essentially describes the dynamics of the equilibrium state of the noninteracting system. Eventually we are interested in a single-particle observable. As already mentioned above, the equilibrium state of the noninteracting system is believed to reasonably describe single-particle observables in equilibrium even for weakly interacting systems. Since an equilibrium state is constant, the above commutator should not significantly contribute to the relevant dynamics; thus we drop it. Keeping the term and

performing all following steps yields eventually an expression which can explicitly be shown to be indeed negligible in the weak-coupling limit. For clarity and briefness we omit this calculation here.

If we apply now Eq. (3) to Eq. (8) and make use of Eq. (9), we obtain

$$\partial_t \mathcal{P}\varrho(t) = \frac{1}{\sigma_{\mathbf{j}}^2}\partial_t d_{\mathbf{j}}(t)\cdot\tilde{\varrho}\otimes\Delta_{\mathbf{j}} = \int_0^t dt'\,\varrho^{\mathrm{eq}} - \int_0^t dt'\frac{1}{\sigma_{\mathbf{j}}^4\hbar^2} \times \mathrm{Tr}\{[\Delta_{\mathbf{j}}[\hat{V}(t),[\hat{V}(t'),\tilde{\varrho}\otimes\Delta_{\mathbf{j}}]]\}\tilde{\varrho}\otimes\Delta_{\mathbf{j}} d_{\mathbf{j}}(t). \tag{10}$$

Multiplying both sides of Eq. (10) with $\Delta_{\mathbf{j}}$ and taking the trace leads to

$$\partial_t d_{\mathbf{j}}(t) = \underbrace{-\frac{1}{\hbar^2\sigma_{\mathbf{j}}^2}\int_0^t dt'\ \mathrm{Tr}\{\Delta_{\mathbf{j}}[\hat{V}(t)[\hat{V}(t'),\tilde{\varrho}\otimes\Delta_{\mathbf{j}}]]\}}_{\Gamma_{\mathbf{j}}(t)} d_{\mathbf{j}}(t), \tag{11}$$

where $\Gamma_{\mathbf{j}}(t)$ appears as a time-dependent damping rate of the mode $\mathbf{j}$. If $\Gamma_{\mathbf{j}}(t)$ turns out to be approximately time independent, the usual exponential relaxation results. With the substitution $t'=t-\tau$ and exploiting $[\tilde{\varrho}\otimes\Delta_{\mathbf{j}},\hat{H}_0]=[\Delta_{\mathbf{j}},\hat{H}_0]=0$ as well as some trace properties, we obtain for the rate:

$$\Gamma_{\mathbf{j}}(t) = \frac{1}{\hbar^2\sigma_{\mathbf{j}}^2}\int_0^{-t} d\tau \underbrace{\mathrm{Tr}\{[\hat{V}(\tau),\tilde{\varrho}\otimes\Delta_{\mathbf{j}}]\cdot[\hat{V}(0),\Delta_{\mathbf{j}}]\}}_{C(\tau)}, \tag{12}$$

where $C(\tau)$ denotes the correlation function which is real due to the fact that both commutators are Hermitian. The concrete evaluation of this expression with respect to our model is straightforward but somewhat lengthy. Thus the full computation is given in the Appendix, here we only give and discuss the results. After exploiting the commutators within the trace, we finally obtain for the rate:

$$\Gamma_{\mathbf{j}}(t,T) = \frac{1}{\tau_{\mathbf{j}}} = -\frac{2}{\hbar^2\sigma_{\mathbf{j}}^2}\sum_{\mathbf{k},\mathbf{q}}\int_0^{-t} d\tau |V(\mathbf{q})|^2 F(\mathbf{k},\mathbf{q},\mathbf{j},T) \times \cos[(\omega_{\mathbf{k}+\mathbf{q}}+\omega_{\mathbf{j}-\mathbf{q}}-\omega_{\mathbf{k}}-\omega_{\mathbf{j}})\tau] = \frac{2}{\hbar^2\sigma_{\mathbf{j}}^2}\sum_{\mathbf{k},\mathbf{q}}|V(\mathbf{q})|^2 F(\mathbf{k},\mathbf{q},\mathbf{j},T)t\ \mathrm{sinc}[\omega(\mathbf{k},\mathbf{q},\mathbf{j})t], \tag{13}$$

with

$$\omega(\mathbf{k},\mathbf{q},\mathbf{j}) := \omega_{\mathbf{k}+\mathbf{q}}+\omega_{\mathbf{j}-\mathbf{q}}-\omega_{\mathbf{k}}-\omega_{\mathbf{j}},$$

$$F(\mathbf{k},\mathbf{q},\mathbf{j},T) := \underbrace{(1-f_{\mathbf{j}})(1-f_{\mathbf{j}-\mathbf{q}})(1-f_{\mathbf{k}+\mathbf{q}})f_{\mathbf{k}}}_{F_1(\mathbf{k},\mathbf{q},\mathbf{j},T)} + \underbrace{f_{\mathbf{j}}f_{\mathbf{j}-\mathbf{q}}f_{\mathbf{k}+\mathbf{q}}(1-f_{\mathbf{k}})}_{F_2(\mathbf{k},\mathbf{q},\mathbf{j},T)}, \tag{14}$$

and $\mathrm{sinc}(\omega t)$ denotes the sinus cardinalis. Obviously the integral $\int_{-\infty}^{+\infty} t\ \mathrm{sinc}(\omega t)d\omega$ is independent of t. Furthermore the function gets more and more peaked with increasing t such that, as well known,

$$\lim_{t\to\infty}\frac{\sin(\omega t)}{\omega} = \pi\delta(\omega). \tag{15}$$

Hence, since dispersion relations are smooth functions of the wave number, we expect the rate $\Gamma_{\mathbf{j}}(t,T)$ to become indeed time independent for times larger than τ_c if $1/\tau_c$ is an energy scale on which dispersion relations may be linearized. Thus for times t larger than τ_c we may with good precision approximate (here we neglect the factor $\sigma_{\mathbf{j}}^{-2}$ since for temperatures $T\approx 0$ K it is $\sigma_{\mathbf{j}}^{-2}\approx 1$):

$$\Gamma_{\mathbf{j}}(T) = \frac{1}{\tau_{\mathbf{j}}} = \frac{2\pi}{\hbar}\sum_{\mathbf{k},\mathbf{q}}|V(\mathbf{q})|^2\delta(\varepsilon_{\mathbf{k}+\mathbf{q}}+\varepsilon_{\mathbf{j}-\mathbf{q}}-\varepsilon_{\mathbf{k}}-\varepsilon_{\mathbf{j}}) \times\{(1-f_{\mathbf{j}})(1-f_{\mathbf{j}-\mathbf{q}})(1-f_{\mathbf{k}+\mathbf{q}})f_{\mathbf{k}}+f_{\mathbf{j}}f_{\mathbf{j}-\mathbf{q}}f_{\mathbf{k}+\mathbf{q}}(1-f_{\mathbf{k}})\}. \tag{16}$$

This expression is one of our main results. In principle it allows for a direct calculation of lifetimes for any fermionic system with given quasiparticle dispersion relations and screened scattering term. Very similar formulas can be found in textbooks in the context of transport and relaxation, see, e.g., Refs. 10 and 11. They are often derived on the basis of an *ad hoc* application of Fermi's golden rule. A closer look reveals that such an expression can also be obtained from an analysis along the lines described in the introduction ("RPA GW") by using the static-limit form of the screened interaction. Since our further quantitative determination only consists in a numerical evaluation of Eq. (16) the outcome is equivalent to the one obtained by the above treatment. Moreover since, as outlined in the following, Eq. (16) is in accord with a standard scattering interpretation, obviously both, the projective and the above version of the many-body approach amount more or less to the counting of scattering events.

The contributions to the decay rate corresponding to F_1 and F_2 allow for an intuitive interpretation, at least for low temperatures.

F_1: this term accounts for the decay of an electron from a momentum mode $\mathbf{j}$ above the Fermi sea, cf. Fig. 1(a). Due to the factor $(1-f_{\mathbf{j}})$ it only significantly contributes to the occupation number dynamics of such modes that are unoccupied in equilibrium. Those occupation numbers may only deviate from equilibrium toward an excess of electrons. According to the other three factors only those summands contribute that correspond to the electron at $\mathbf{j}$ colliding with an electron from within the Fermi sea $\mathbf{k}$, such that the postcol-

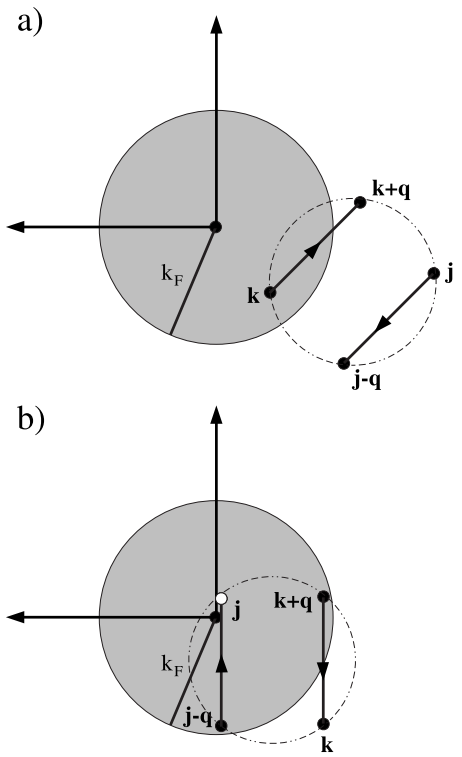

FIG. 1. Schematic representation of the underlying collision processes in momentum space as described in the text (k_F denotes the Fermi momentum). (a) A collision process through which an excited electron at momentum **j** vanishes from its initial momentum mode. (b) A collision process "filling" a hole within the Fermi sphere at **j**. The dashed circles denote the possible outgoing momenta under momentum and energy conservations.

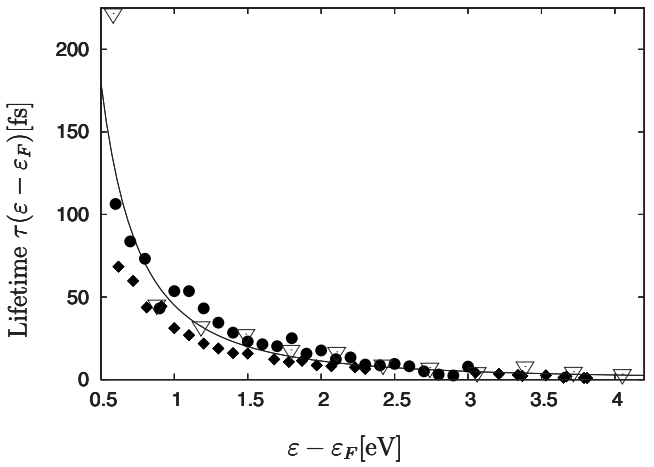

FIG. 2. Comparison of the lifetimes of excited electrons in aluminum as arising from electron-electron scattering only. Displayed is the regime close to the Fermi edge. The open triangles correspond to data obtained from the approach at hand, i.e., integration of Eq. (22) ($T=10$ K). The solid circles are experimental data corrected for transport effects taken from Ref. 4 while the solid line is the analytical result from Ref. 1. Solid diamonds are the results of an DFT-GW calculation as shown in Ref. 8. The number of sample points for the Monte Carlo integration of Eq. (22) is $N=2\times10^7$, the broadening parameter is $\sigma=1/25$.

lision momenta $\mathbf{k}+\mathbf{q}$ and $\mathbf{j}-\mathbf{q}$ lay in the unoccupied region above the Fermi sea.

F_2: this term accounts for the decay of a hole from a momentum mode **j** within the Fermi sea, cf. Fig. 1(b). Due to the factor $f_\mathbf{j}$ it only significantly contributes to the occupation number dynamics of such modes that are occupied in equilibrium. Those occupation numbers may only deviate from equilibrium toward a shortage of electrons, i.e., holes. According to the other three factors only those summands contribute that correspond to two electrons from within the Fermi sea $\mathbf{k}+\mathbf{q}$ and $\mathbf{j}-\mathbf{q}$ colliding such that one postcollision momentum **k** lies in the unoccupied region above the Fermi sea and the other lays exactly at **j** such as to fill up the hole.

III. APPLICATION TO A JELLIUM MODEL WITH SCREENED INTERACTION

In this section we now apply our result for the decay rates to a jellium model featuring a Thomas-Fermi screened interaction. The latter will eventually be tuned to correspond to aluminum. However, to repeat, the main intention of this work is not to calculate decay rates in aluminum with extreme precision but to concretely demonstrate the feasibility of our method. The Hamiltonian of the model is given by

$$\hat{H}_J=\frac{\hbar^2}{2m_e}\sum_{\mathbf{k}}k^2a_\mathbf{k}^\dagger a_\mathbf{k}+\frac{1}{2}\sum_{\mathbf{k},\mathbf{l},\mathbf{q}}\frac{e^2}{\Omega\varepsilon_0(\mathbf{q}^2+\mathbf{q}_{\mathrm{TF}}^2)}a_{\mathbf{k}+\mathbf{q}}^\dagger a_{\mathbf{l}-\mathbf{q}}^\dagger a_\mathbf{l}a_\mathbf{k}, \tag{17}$$

where Ω denotes the volume of the solid and $\mathbf{q}_{\mathrm{TF}}$ the so-called Thomas-Fermi wave number which is related to the Fermi wave vector and the Wigner-Seitz radius by $(q_{\mathrm{TF}}/k_F)^2=0.665r_S$ [with $r_S=(\frac{3}{4\pi\rho_0})^{1/3}\frac{1}{a_0}$, a_0 being the Bohr radius, $\varepsilon_F=(9\pi/4)^{2/3}\frac{1}{r_S^2}$ [Ry], $k_F=(9\pi/4)^{1/3}\frac{1}{a_0r_S}$, and $q_{\mathrm{TF}}=(12/\pi)^{1/3}\frac{1}{a_0\sqrt{r_S}}$]. Note that our model only comprises electrons, no phonons. Thus the result on the decay rate has to be compared to that part of the total decay rate that stems from electron-electron scattering only. In real aluminum there is evidence that the total lifetime is also significantly shortened due to electron-phonon scattering.[4,27] We apply now Eq. (16) to this model which yields

$$\Gamma_\mathbf{j}(T)=\frac{1}{\tau_\mathbf{j}}=\frac{4\pi}{\hbar}\sum_{\mathbf{k},\mathbf{q}}\left[\frac{e^2}{\Omega\varepsilon_0(\mathbf{q}^2+\mathbf{q}_{\mathrm{TF}}^2)}\right]^2F(\mathbf{k},\mathbf{q},\mathbf{j},T)\times\delta(\varepsilon_{\mathbf{k}+\mathbf{q}}+\varepsilon_{\mathbf{j}-\mathbf{q}}-\varepsilon_\mathbf{k}-\varepsilon_\mathbf{j}), \tag{18}$$

where the auxiliary factor of 2 arises from the fact that for each **k** we have two one-electron states (one for each spin) which is taken into account by this additional factor. If we now replace the sums in Eq. (18) by integrals by the rule $\Sigma_\mathbf{k}f(\mathbf{k})\to(2\pi)^{-3}\Omega\int d\mathbf{k}f(\mathbf{k})$, we obtain for $\Gamma_\mathbf{j}(T)$:

$$\Gamma_{\mathbf{j}}(T) = \frac{4\Omega^2}{2^6\hbar\pi^5}\int\int d\mathbf{k}d\mathbf{q}\left(\frac{e^2}{\Omega\varepsilon_0(\mathbf{q}^2+\mathbf{q}_{\mathrm{TF}}^2)}\right)^2 F(\mathbf{k},\mathbf{q},\mathbf{j},T) \times \delta(\varepsilon_{\mathbf{k}+\mathbf{q}}+\varepsilon_{\mathbf{j}-\mathbf{q}}-\varepsilon_{\mathbf{k}}-\varepsilon_{\mathbf{j}}) = \frac{e^4}{16\hbar\varepsilon_0^2\pi^5}\int\int d\mathbf{k}d\mathbf{q}\frac{F(\mathbf{k},\mathbf{q},\mathbf{j},T)}{(\mathbf{q}^2+\mathbf{q}_{\mathrm{TF}}^2)^2} \times \delta(\varepsilon_{\mathbf{k}+\mathbf{q}}+\varepsilon_{\mathbf{j}-\mathbf{q}}-\varepsilon_{\mathbf{k}}-\varepsilon_{\mathbf{j}}). \quad (19)$$

For the numerical calculation it is advantageous to transform the momenta to dimensionless parameter; thus we introduce coordinates relative to the Fermi momentum: $\mathbf{k}\rightarrow k_F\cdot\mathbf{k}'$, thus $\mathbf{q}\rightarrow k_F\cdot\mathbf{q}'$ and thus $\mathbf{j}\rightarrow k_F\cdot\mathbf{j}'$.

Applying all these substitutions to Eq. (19) leads to

$$\Gamma_{\mathbf{j}'}(T) = \frac{m_e e^4}{8\hbar^3\varepsilon_0^2\pi^5}\int\int d\mathbf{k}'d\mathbf{q}'\frac{F(\mathbf{k}',\mathbf{q}',\mathbf{j}',T)}{(\mathbf{q}'^2+0.665r_S)^2} \times \delta[(\mathbf{k}'+\mathbf{q}')^2+(\mathbf{j}'-\mathbf{q}')^2-\mathbf{k}'^2-\mathbf{j}'^2], \quad (20)$$

with $f_{\mathbf{k}'}=\{\exp[\beta\varepsilon_F(\mathbf{k}'^2-1)]+1\}^{-1}$ and $m_e=9.1\times10^{-31}$ kg for the free-electron mass.

For the numerical evaluation of the last expression we approximate the delta distribution by a suitable nonsingular, e.g., Gaussian-type function:

$$\delta_\sigma(\omega) \approx \frac{1}{\sigma\sqrt{2\pi}}e^{-(\omega^2/2\sigma^2)}, \quad (21)$$

where σ denotes the standard deviation which has to be in some sense small. More details on this somewhat subtle approximation are given below. Thus we get the following integral for the rate:

$$\Gamma_{\mathbf{j}'}(T) = \frac{1.05\ \mathrm{fs}^{-1}}{\sigma}\cdot\int\int d\mathbf{k}'d\mathbf{q}'\frac{F(\mathbf{k}',\mathbf{q}',\mathbf{j}',T)}{(\mathbf{q}'^2+0.665r_S)^2} \times \exp\left\{\frac{1}{2\sigma^2}[(\mathbf{k}'+\mathbf{q}')^2+(\mathbf{j}'-\mathbf{q}')^2-\mathbf{k}'^2-\mathbf{j}'^2]^2\right\}. \quad (22)$$

For aluminum we choose $r_S=2.07$. The six dimensional integrals are solved numerically without any further simplification using a standard Monte Carlo package as implemented in the MATHEMATICA code. Of course this specific integral could be evaluated in other ways; however, to demonstrate the feasibility of our approach in general we proceed as indicated.

As one can see in Fig. 2 there is rather good agreement between our results, other theoretical approaches, and experiment. Our data is denoted by open triangles. The solid line corresponds to the many-body approach based on jellium[1] as outlined in Sec. I. The solid diamonds denote the result of a more sophisticated many-body approach which takes the lattice into account and exploits density-functional theory.[8] The solid circles indicate the parts of the measured decay rates that are attributed to direct electron-electron scattering, i.e., after removal of transport effects according.[4] Figure 3 shows the analytical result from[1] $[263r_S^{-5/2}(\varepsilon-\varepsilon_F)^{-2}\ [\mathrm{eV}]^2\ [\mathrm{fs}]]$, which is supposed to be valid close to the Fermi edge, boldly continued to all energies (solid line). Furthermore results of our approach for all energies are displayed (dots). Obviously there are deviations for electrons at higher energies while the agreement remains very good in the limit of "low-energy holes."

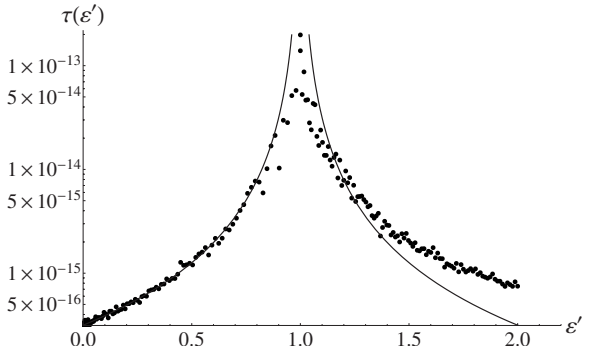

FIG. 3. Comparison of the logarithmic lifetimes of excited electrons (above $\varepsilon'=1$) and holes (below $\varepsilon'=1$) in aluminum as arising from electron-electron scattering only. Displayed is a wide regime around the Fermi edge. Data are plotted over rescaled energy, $\varepsilon'=\varepsilon/\varepsilon_F$. Displayed are results obtained from Fermi-liquid theory as cited in Ref. 1 (solid line, $T=0$ K) and from numerical integration of Eq. (22) (dots, $T=10$ K). The number of sample points for the Monte Carlo integration of Eq. (22) is $N=10^7$ and $\sigma=1/10$.

However, a comment should be added here. For this more or less realistic model we get lifetimes on the order of some femtoseconds. The decay time of correlation function (12) is, very roughly, on the order of $h/\epsilon_{\max}$, with $\epsilon_{\max}$ being the bandwidth. For about 10 eV this yields ca. half a femtosecond. Thus the separation of those time scales, which has been mentioned in Sec. III as a criterion for the truncation performed above, is not as clear as often in other fields, such as, e.g., quantum optics. This indicates that such models, at short lifetimes, are barely in the Markovian weak-coupling regime, and hence memory effects and/or higher orders may have significant influence.

To the choice of σ: Obviously a smaller σ leads to a better approximation of the δ function which should be the correct weight distribution at least in the long-time limit. However, recall the above discussion of the time independence of the decay rate. For analogous reasons larger σ should leave the result unaltered, as long as σ remains small enough to allow for a linearization of the dispersion relations on the scale of σ. A large σ is numerically favorable since the larger σ is, the larger will be the fraction of the Monte Carlo points that significantly contribute to the integral. Moreover of course this yields a decreasing statistical error. Thus, for a given statistical integration error, a larger σ simply implies a longer computing time. Hence finding the best σ is an optimization process that should be done carefully. However, to name a number, the computation time for one of the lifetimes as displayed in Figs. 2 and 3 is about an hour.

IV. SUMMARY, CONCLUSION, AND OUTLOOK

In this paper we considered the lifetimes of (quasi)particles or holes in interacting quantum gases (only electronic part), using a projection operator technique. This yields a formula for the decay rates into which essentially the pertinent effective quasiparticle dispersion relations of the particles and their screened interactions enter. This formula turns out to be in accord with an expression that may be found from a certain implementation of the self-energy formalism. The rates are eventually given in terms of integrals which can be cast into a form which is well suited for a Monte Carlo integration scheme. While this work essentially aims at demonstrating the feasibility of this approach in general, the method has been concretely applied to a jellium model featuring a Thomas-Fermi screened interaction (tuned for aluminum) as a simple example. Here it yields reasonable results while requiring moderate computational effort. This motivates an application of the approach to more complex systems. However, the results on life and correlation times indicate that such systems are, for short lifetimes (high electron energies, etc.), barely Markovian and thus the decay may not even be strictly exponential. This hints at a necessity to include higher order terms in future investigations in this regime.

The approach at hand aimed at generating an autonomous linear equation of motion for single electron occupation number (11). However a slight modification of the projection used here may directly yield linear equation of motion for all electron occupation numbers, i.e., a linearized Boltzmann equation. As well known, the latter is a traditional starting point in investigating, e.g., transport properties. To those ends one would use a projection very much like the one discussed here [Eq. (8)] but summed over all occupation numbers j. The reasonable results on lifetimes presented in this work may be viewed to encourage further investigations in that direction.

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APPENDIX: DERIVATION OF THE DECAY RATE

In this section we show the derivation of Eq. (16). The main work is to exploit the two commutators and finally the trace. First we exploit the commutator $[\hat{V}(0),\Delta_{\mathbf{j}}]$:

$$[\hat{V}(0),\Delta_{\mathbf{j}}] = \frac{1}{2}\sum_{\mathbf{k},\mathbf{l},\mathbf{q}} V(\mathbf{q})[a^\dagger_{\mathbf{k}+\mathbf{q}}a^\dagger_{\mathbf{l}-\mathbf{q}}a_{\mathbf{l}}a_{\mathbf{k}}, a^\dagger_{\mathbf{j}}a_{\mathbf{j}} - f_{\mathbf{j}}]$$

$$= \frac{1}{2}\sum_{\mathbf{k},\mathbf{l},\mathbf{q}} V(\mathbf{q})[a^\dagger_{\mathbf{k}+\mathbf{q}}a^\dagger_{\mathbf{l}-\mathbf{q}}a_{\mathbf{l}}a_{\mathbf{k}}, a^\dagger_{\mathbf{j}}a_{\mathbf{j}}].$$

Since the commutator is zero for $\mathbf{j}\neq\mathbf{k}+\mathbf{q},\mathbf{l}-\mathbf{q},\mathbf{k},\mathbf{l}$, we just have to regard cases where one of the indices is equal to j and note that $a_{\mathbf{i}}a^\dagger_{\mathbf{i}}a_{\mathbf{i}}=a_{\mathbf{i}}$ and $a^\dagger_{\mathbf{i}}a^\dagger_{\mathbf{i}}a_{\mathbf{i}}=0$. From this follows that

$$[\hat{V}(0),\Delta_{\mathbf{j}}] = \frac{1}{2}\sum_{\mathbf{k},\mathbf{q}} V(\mathbf{q})a^\dagger_{\mathbf{k}+\mathbf{q}}a^\dagger_{\mathbf{j}-\mathbf{q}}a_{\mathbf{k}}a_{\mathbf{j}} - \frac{1}{2}\sum_{\mathbf{l},\mathbf{q}} V(\mathbf{q})a^\dagger_{\mathbf{j}+\mathbf{q}}a^\dagger_{\mathbf{l}-\mathbf{q}}a_{\mathbf{l}}a_{\mathbf{j}}$$
$$- \frac{1}{2}\sum_{\mathbf{k},\mathbf{q}} V(\mathbf{q})a^\dagger_{\mathbf{j}}a^\dagger_{\mathbf{k}}a_{\mathbf{j}-\mathbf{q}}a_{\mathbf{k}+\mathbf{q}}$$
$$+ \frac{1}{2}\sum_{\mathbf{l},\mathbf{q}} V(\mathbf{q})a^\dagger_{\mathbf{j}}a^\dagger_{\mathbf{l}}a_{\mathbf{l}-\mathbf{q}}a_{\mathbf{j}+\mathbf{q}}. \quad \text{(A1)}$$

With suitable index shifts and the fermionic commutator relations, we finally obtain for the commutator

$$[\hat{V}(0),\Delta_{\mathbf{j}}] = \sum_{\mathbf{k},\mathbf{q}} V(\mathbf{q})(a^\dagger_{\mathbf{k}+\mathbf{q}}a^\dagger_{\mathbf{j}-\mathbf{q}}a_{\mathbf{k}}a_{\mathbf{j}} - a^\dagger_{\mathbf{j}}a^\dagger_{\mathbf{k}}a_{\mathbf{j}-\mathbf{q}}a_{\mathbf{k}+\mathbf{q}}). \quad \text{(A2)}$$

Now we deal with the second commutator $[\hat{V}(\tau),\tilde{\varrho}\otimes\Delta_{\mathbf{j}}]$ where we first regard the case $\mathbf{j}\neq\mathbf{k}+\mathbf{q},\mathbf{l}-\mathbf{q},\mathbf{k},\mathbf{l}$ (we abbreviate $g_{\mathbf{i}}:=1-f_{\mathbf{i}}$):

$$[\hat{V}(\tau),\tilde{\varrho}\otimes\Delta_{\mathbf{j}}]^{\neq\mathbf{j}}$$
$$= \frac{1}{2}\sum_{\mathbf{k},\mathbf{l},\mathbf{q}} V(\mathbf{q})[a^\dagger_{\mathbf{k}+\mathbf{q}}(\tau)a^\dagger_{\mathbf{l}-\mathbf{q}}(\tau)a_{\mathbf{l}}(\tau)a_{\mathbf{k}}(\tau),\tilde{\varrho}_s\otimes\Delta_{\mathbf{j}}]$$
$$= \frac{1}{2}\sum_{\mathbf{k},\mathbf{l},\mathbf{q}} V(\mathbf{q})e^{(\imath/\hbar)(\varepsilon_{\mathbf{k}+\mathbf{q}}+\varepsilon_{\mathbf{l}-\mathbf{q}}-\varepsilon_{\mathbf{k}}-\varepsilon_{\mathbf{l}})\tau}$$
$$\times[a^\dagger_{\mathbf{k}+\mathbf{q}}a^\dagger_{\mathbf{l}-\mathbf{q}}a_{\mathbf{l}}a_{\mathbf{k}}, \bigotimes_{\mathbf{i}\neq\mathbf{j}}(f_{\mathbf{i}}a^\dagger_{\mathbf{i}}a_{\mathbf{i}} + g_{\mathbf{i}}a_{\mathbf{i}}a^\dagger_{\mathbf{i}})\otimes\Delta_{\mathbf{j}}],$$

where the annihilation and creation operators act only on the respective subspaces of the tensor product of the single density operators. With $a_{\mathbf{i}}a_{\mathbf{i}}a^\dagger_{\mathbf{i}}=0$, $a^\dagger_{\mathbf{i}}a_{\mathbf{i}}a^\dagger_{\mathbf{i}}=a^\dagger_{\mathbf{i}}$, the rules above, and $u(\tau):=\exp[\frac{\imath}{\hbar}(\varepsilon_{\mathbf{k}+\mathbf{q}}+\varepsilon_{\mathbf{l}-\mathbf{q}}-\varepsilon_{\mathbf{k}}-\varepsilon_{\mathbf{l}})\tau]$ it follows:

$$[\hat{V}(\tau),\tilde{\varrho}\otimes\Delta_{\mathbf{j}}]^{\neq\mathbf{j}} = \frac{1}{2}\sum_{\mathbf{k},\mathbf{l},\mathbf{q}} V(\mathbf{q})u(\tau)$$
$$\times(f_{\mathbf{k}}f_{\mathbf{l}}g_{\mathbf{l}-\mathbf{q}}g_{\mathbf{k}+\mathbf{q}} - g_{\mathbf{k}}g_{\mathbf{l}}f_{\mathbf{l}-\mathbf{q}}f_{\mathbf{k}+\mathbf{q}})$$
$$\times a^\dagger_{\mathbf{k}+\mathbf{q}}a^\dagger_{\mathbf{l}-\mathbf{q}}a_{\mathbf{l}}a_{\mathbf{k}}\bigotimes_{\substack{\mathbf{i}\neq\mathbf{j},\mathbf{k},\\ \mathbf{l},\mathbf{k}+\mathbf{q},\\ \mathbf{l}-\mathbf{q}}}\varrho^{\mathrm{eq}}_{\mathbf{i}}\otimes\Delta_{\mathbf{j}}. \quad \text{(A3)}$$

For the cases where one of the indices $\mathbf{k}+\mathbf{q},\mathbf{l}-\mathbf{q},\mathbf{k},\mathbf{l}$ is equal to $\mathbf{j}$, we obtain analogous

$$
\begin{aligned}
[\hat{V}(\tau),\tilde{\varrho}\otimes\Delta_{\mathbf{j}}]^{=\mathbf{j}} &= \frac{1}{2}\sum_{\mathbf{k},\mathbf{q}} V(\mathbf{q})a^{\dagger}_{\mathbf{k}+\mathbf{q}}(\tau)a^{\dagger}_{\mathbf{j}-\mathbf{q}}(\tau)a_{\mathbf{k}}(\tau)a_{\mathbf{j}}(\tau)\bigotimes_{\substack{\mathbf{i}\neq\mathbf{j},\mathbf{k},\\ \mathbf{j}-\mathbf{q},\\ \mathbf{k}+\mathbf{q}}}\varrho^{\mathrm{eq}}_{\mathbf{i}}(g_{\mathbf{j}}g_{\mathbf{j}-\mathbf{q}}g_{\mathbf{k}+\mathbf{q}}f_{\mathbf{k}}+f_{\mathbf{j}}f_{\mathbf{j}-\mathbf{q}}f_{\mathbf{k}+\mathbf{q}}g_{\mathbf{k}})\\
&+\frac{1}{2}\sum_{\mathbf{l},\mathbf{q}} V(\mathbf{q})a^{\dagger}_{\mathbf{j}+\mathbf{q}}(\tau)a^{\dagger}_{\mathbf{l}-\mathbf{q}}(\tau)a_{\mathbf{j}}(\tau)a_{\mathbf{l}}(\tau)\bigotimes_{\substack{\mathbf{i}\neq\mathbf{j},\mathbf{l},\\ \mathbf{l}-\mathbf{q},\\ \mathbf{j}+\mathbf{q}}}\varrho^{\mathrm{eq}}_{\mathbf{i}}(g_{\mathbf{j}}g_{\mathbf{l}-\mathbf{q}}g_{\mathbf{j}+\mathbf{q}}f_{\mathbf{l}}+f_{\mathbf{j}}f_{\mathbf{j}+\mathbf{q}}f_{\mathbf{l}-\mathbf{q}}g_{\mathbf{l}})\\
&-\frac{1}{2}\sum_{\mathbf{k},\mathbf{q}} V(\mathbf{q})a^{\dagger}_{\mathbf{k}+\mathbf{q}}(\tau)a^{\dagger}_{\mathbf{j}}(\tau)a_{\mathbf{k}}(\tau)a_{\mathbf{j}+\mathbf{q}}(\tau)\bigotimes_{\substack{\mathbf{i}\neq\mathbf{j},\mathbf{j}+\mathbf{q},\\ \mathbf{k},\mathbf{k}+\mathbf{q}}}\varrho^{\mathrm{eq}}_{\mathbf{i}}(g_{\mathbf{j}+\mathbf{q}}g_{\mathbf{k}}g_{\mathbf{j}}f_{\mathbf{k}+\mathbf{q}}+f_{\mathbf{j}}f_{\mathbf{k}}f_{\mathbf{j}+\mathbf{q}}g_{\mathbf{k}+\mathbf{q}})\\
&-\frac{1}{2}\sum_{\mathbf{l},\mathbf{q}} V(\mathbf{q})a^{\dagger}_{\mathbf{j}}(\tau)a^{\dagger}_{\mathbf{l}-\mathbf{q}}(\tau)a_{\mathbf{j}-\mathbf{q}}(\tau)a_{\mathbf{l}}(\tau)\bigotimes_{\substack{\mathbf{i}\neq\mathbf{j},\mathbf{j}-\mathbf{q},\\ \mathbf{l},\mathbf{l}-\mathbf{q}}}\varrho^{\mathrm{eq}}_{\mathbf{i}}(g_{\mathbf{j}-\mathbf{q}}g_{\mathbf{l}}g_{\mathbf{j}}f_{\mathbf{l}-\mathbf{q}}+f_{\mathbf{j}}f_{\mathbf{l}}f_{\mathbf{j}-\mathbf{q}}g_{\mathbf{l}-\mathbf{q}}).
\end{aligned}
\tag{A4}
$$

Again with suitable index shifts and substitutions it follows for the commutator:

$$
[\hat{V}(\tau),\tilde{\varrho}\otimes\Delta_{\mathbf{j}}]^{=\mathbf{j}} = \sum_{\mathbf{k},\mathbf{q}} V(\mathbf{q})[a^{\dagger}_{\mathbf{k}+\mathbf{q}}(\tau)a^{\dagger}_{\mathbf{j}-\mathbf{q}}(\tau)a_{\mathbf{k}}(\tau)a_{\mathbf{j}}(\tau)-a^{\dagger}_{\mathbf{j}}(\tau)a^{\dagger}_{\mathbf{k}}(\tau)a_{\mathbf{j}-\mathbf{q}}(\tau)a_{\mathbf{k}+\mathbf{q}}(\tau)](g_{\mathbf{j}}g_{\mathbf{j}-\mathbf{q}}g_{\mathbf{k}+\mathbf{q}}f_{\mathbf{k}}+f_{\mathbf{j}}f_{\mathbf{j}-\mathbf{q}}f_{\mathbf{k}+\mathbf{q}}g_{\mathbf{k}})\bigotimes_{\substack{\mathbf{i}\neq\mathbf{j},\mathbf{k},\\ \mathbf{j}-\mathbf{q},\\ \mathbf{k}+\mathbf{q}}}\varrho^{\mathrm{eq}}_{\mathbf{i}}.
\tag{A5}
$$

Now we exploit the trace:

$$
\begin{aligned}
C(\tau) &= \mathrm{Tr}\{[\hat{V}(\tau),\tilde{\varrho}\otimes\Delta_{\mathbf{j}}][\hat{V}(0),\Delta_{\mathbf{j}}]\} = \mathrm{Tr}\{([\hat{V}(\tau),\tilde{\varrho}\otimes\Delta_{\mathbf{j}}]^{\neq\mathbf{j}}+[\hat{V}(\tau),\tilde{\varrho}\otimes\Delta_{\mathbf{j}}]^{=\mathbf{j}})[\hat{V}(0),\Delta_{\mathbf{j}}]\}\\
&= \underbrace{\mathrm{Tr}\{[\hat{V}(\tau),\tilde{\varrho}\otimes\Delta_{\mathbf{j}}]^{\neq\mathbf{j}}[\hat{V}(0),\Delta_{\mathbf{j}}]\}}_{A(\tau)} + \underbrace{\mathrm{Tr}\{[\hat{V}(\tau),\tilde{\varrho}\otimes\Delta_{\mathbf{j}}]^{=\mathbf{j}}[\hat{V}(0),\Delta_{\mathbf{j}}]\}}_{B(\tau)},
\end{aligned}
$$

where we split it into two parts and exploit them, respectively:

$$
\begin{aligned}
A(\tau) &= \frac{1}{2}\sum_{\substack{\mathbf{k},\mathbf{l},\mathbf{q}\\ \mathbf{x},\mathbf{y}}} V(\mathbf{q})V(\mathbf{y})u(\tau)G(\mathbf{k},\mathbf{l},\mathbf{q},T)\mathrm{Tr}\left\{a^{\dagger}_{\mathbf{k}+\mathbf{q}}a^{\dagger}_{\mathbf{l}-\mathbf{q}}a_{\mathbf{l}}a_{\mathbf{k}}\bigotimes_{\substack{\mathbf{i}\neq\mathbf{j},\mathbf{k},\\ \mathbf{l},\mathbf{k}+\mathbf{q},\\ \mathbf{l}-\mathbf{q}}}\varrho^{\mathrm{eq}}_{\mathbf{i}}\otimes\Delta_{\mathbf{j}}(a^{\dagger}_{\mathbf{x}+\mathbf{y}}a^{\dagger}_{\mathbf{j}-\mathbf{y}}a_{\mathbf{x}}a_{\mathbf{j}}-a^{\dagger}_{\mathbf{j}}a^{\dagger}_{\mathbf{x}}a_{\mathbf{j}-\mathbf{y}}a_{\mathbf{x}+\mathbf{y}})\right\}\\
&= \frac{1}{2}\sum_{\substack{\mathbf{k},\mathbf{l},\mathbf{q}\\ \mathbf{x},\mathbf{y}}} V(\mathbf{q})V(\mathbf{y})u(\tau)G(\mathbf{k},\mathbf{l},\mathbf{q},T)\underbrace{\mathrm{Tr}\left\{a^{\dagger}_{\mathbf{k}+\mathbf{q}}a^{\dagger}_{\mathbf{l}-\mathbf{q}}a_{\mathbf{l}}a_{\mathbf{k}}\bigotimes_{\substack{\mathbf{i}\neq\mathbf{j},\mathbf{k},\\ \mathbf{l},\mathbf{k}+\mathbf{q},\\ \mathbf{l}-\mathbf{q}}}\varrho^{\mathrm{eq}}_{\mathbf{i}}\otimes\Delta_{\mathbf{j}}a^{\dagger}_{\mathbf{x}+\mathbf{y}}a^{\dagger}_{\mathbf{j}-\mathbf{y}}a_{\mathbf{x}}a_{\mathbf{j}}\right\}}_{\mathrm{I}}\\
&\quad -\frac{1}{2}\sum_{\substack{\mathbf{k},\mathbf{l},\mathbf{q}\\ \mathbf{x},\mathbf{y}}} V(\mathbf{q})V(\mathbf{y})u(\tau)G(\mathbf{k},\mathbf{l},\mathbf{q},T)\underbrace{\mathrm{Tr}\left\{a^{\dagger}_{\mathbf{k}+\mathbf{q}}a^{\dagger}_{\mathbf{l}-\mathbf{q}}a_{\mathbf{l}}a_{\mathbf{k}}\bigotimes_{\substack{\mathbf{i}\neq\mathbf{j},\mathbf{k},\\ \mathbf{l},\mathbf{k}+\mathbf{q},\\ \mathbf{l}-\mathbf{q}}}\varrho^{\mathrm{eq}}_{\mathbf{i}}\otimes\Delta_{\mathbf{j}}a^{\dagger}_{\mathbf{j}}a^{\dagger}_{\mathbf{x}}a_{\mathbf{j}-\mathbf{y}}a_{\mathbf{x}+\mathbf{y}}\right\}}_{\mathrm{II}},
\end{aligned}
\tag{A6}
$$

with $G(\mathbf{k},\mathbf{l},\mathbf{q},T)=f_{\mathbf{k}}f_{\mathbf{l}}g_{\mathbf{l}-\mathbf{q}}g_{\mathbf{k}+\mathbf{q}}-g_{\mathbf{k}}g_{\mathbf{l}}f_{\mathbf{l}-\mathbf{q}}f_{\mathbf{k}+\mathbf{q}}$. We focus now on the traces. For taking the trace we use the occupation number representation.

Analysis of part $\underline{\mathbf{I}}$ of $A(\tau)$:

$$\mathrm{Tr}\left\{ a^\dagger_{\mathbf{k+q}} a^\dagger_{\mathbf{l-q}} a_{\mathbf{l}} a_{\mathbf{k}} \underbrace{\bigotimes_{\substack{\mathbf{i}\neq\mathbf{j},\mathbf{k},\\ \mathbf{l},\mathbf{k+q},\\ \mathbf{l-q}}} \varrho^{eq}_{\mathbf{i}}}_{\Xi} \otimes \Delta_{\mathbf{j}} a^\dagger_{\mathbf{x+y}} a^\dagger_{\mathbf{j-y}} a_{\mathbf{x}} a_{\mathbf{j}} \right\} = \sum_{n_1,\ldots,n_r} \langle n_1,\ldots,n_r | a^\dagger_{\mathbf{k+q}} a^\dagger_{\mathbf{l-q}} a_{\mathbf{l}} a_{\mathbf{k}} \cdot \Xi \otimes \Delta_{\mathbf{j}} \cdot a^\dagger_{\mathbf{x+y}} a^\dagger_{\mathbf{j-y}} a_{\mathbf{x}} a_{\mathbf{j}} | n_1,\ldots,n_r \rangle . \tag{A7}$$

Since under consideration no one of the indices $\mathbf{k},\mathbf{l},\mathbf{k+q},\mathbf{l-q}$ is equal to $\mathbf{j}$, this trace is zero for cases with $\mathbf{y}\neq 0$ which is valid for II also. The case $\mathbf{y}=0$ must be analyzed independently.

$\underline{\mathbf{I}}$ for $\mathbf{y}=0$ (now we write down the sum again) we have

$$\frac{1}{2}\sum_{\substack{\mathbf{k},\mathbf{l}\\ \mathbf{q},\mathbf{x}}} V(\mathbf{q})V(0)u(\tau)G(\mathbf{k},\mathbf{l},\mathbf{q},T)\mathrm{Tr}\{a^\dagger_{\mathbf{k+q}} a^\dagger_{\mathbf{l-q}} a_{\mathbf{l}} a_{\mathbf{k}} \cdot \Xi \otimes \Delta_{\mathbf{j}} \cdot a^\dagger_{\mathbf{x}} a^\dagger_{\mathbf{j}} a_{\mathbf{x}} a_{\mathbf{j}}\}. \tag{A8}$$

Here we have two summands: the case where $\mathbf{k+q}=\mathbf{l}$ and $\mathbf{l+q}=\mathbf{l}\rightarrow\mathbf{q}=0$:

$$\begin{aligned}
&\frac{1}{2}\sum_{\substack{\mathbf{k},\mathbf{l}\\ \mathbf{q},\mathbf{x}}} V(\mathbf{q})V(0)u(\tau)G(\mathbf{k},\mathbf{l},\mathbf{q},T)\mathrm{Tr}\{a^\dagger_{\mathbf{k+q}} a^\dagger_{\mathbf{l-q}} a_{\mathbf{l}} a_{\mathbf{k}} \cdot \Xi \otimes \Delta_{\mathbf{j}} \cdot a^\dagger_{\mathbf{x}} a^\dagger_{\mathbf{j}} a_{\mathbf{x}} a_{\mathbf{j}}\} \\
&= \frac{1}{2}\sum_{\mathbf{k},\mathbf{q},\mathbf{x}} V(\mathbf{q})V(0)u(\tau)G(\mathbf{k},\mathbf{k+q},\mathbf{q},T) \\
&\quad \times \mathrm{Tr}\{a^\dagger_{\mathbf{k+q}} a^\dagger_{\mathbf{k}} a_{\mathbf{k+q}} a_{\mathbf{k}} \cdot \Xi \otimes \Delta_{\mathbf{j}} \cdot a^\dagger_{\mathbf{x}} a^\dagger_{\mathbf{j}} a_{\mathbf{x}} a_{\mathbf{j}}\} \\
&\quad + \frac{1}{2}\sum_{\mathbf{k},\mathbf{l},\mathbf{x}} V(0)V(0)u(\tau)G(\mathbf{k},\mathbf{l},\mathbf{0},T)\mathrm{Tr}\{a^\dagger_{\mathbf{k}} a^\dagger_{\mathbf{l}} a_{\mathbf{l}} a_{\mathbf{k}} \cdot \Xi \otimes \Delta_{\mathbf{j}} \cdot a^\dagger_{\mathbf{x}} a^\dagger_{\mathbf{j}} a_{\mathbf{x}} a_{\mathbf{j}}\} = 0,
\end{aligned} \tag{A9}$$

since $G(\mathbf{k},\mathbf{k+q},\mathbf{q},T)=G(\mathbf{k},\mathbf{l},\mathbf{0},T)=0$. For II the argumentation is analogous. Thus there is left just one more possibility: the case $\mathbf{j}=\mathbf{x+y}$ for which we obtain from Eq. (A6):

$$\begin{aligned}
&\frac{1}{2}\sum_{\mathbf{k},\mathbf{l},\mathbf{q},\mathbf{x}} V(\mathbf{q})V(\mathbf{y})u(\tau)G(\mathbf{k},\mathbf{l},\mathbf{q},T)\mathrm{Tr}\{a^\dagger_{\mathbf{k+q}} a^\dagger_{\mathbf{l-q}} a_{\mathbf{l}} a_{\mathbf{k}} \cdot \Xi \\
&\quad \otimes \Delta_{\mathbf{j}} \cdot a^\dagger_{\mathbf{j}} a^\dagger_{\mathbf{j-y}} a_{\mathbf{j-y}} a_{\mathbf{j}}\} - \frac{1}{2}\sum_{\mathbf{k},\mathbf{l},\mathbf{q},\mathbf{x}} V(\mathbf{q})V(\mathbf{y})u(\tau)G(\mathbf{k},\mathbf{l},\mathbf{q},T) \\
&\quad \times \mathrm{Tr}\{a^\dagger_{\mathbf{k+q}} a^\dagger_{\mathbf{l-q}} a_{\mathbf{l}} a_{\mathbf{k}} \cdot \Xi \otimes \Delta_{\mathbf{j}} \cdot a^\dagger_{\mathbf{j}} a^\dagger_{\mathbf{j-y}} a_{\mathbf{j-y}} a_{\mathbf{j}}\} = 0,
\end{aligned} \tag{A10}$$

so that it finally follows that $A(\tau)=0$.

For $B(\tau)$ we have

$$\begin{aligned}
B(\tau) &= \sum_{\substack{\mathbf{k},\mathbf{q}\\ \mathbf{x},\mathbf{y}}} V(\mathbf{q})V(\mathbf{y})F(\mathbf{k},\mathbf{q},\mathbf{j},T)\mathrm{Tr}\{[a^\dagger_{\mathbf{k+q}}(\tau)a^\dagger_{\mathbf{j-q}}(\tau)a_{\mathbf{k}}(\tau)a_{\mathbf{j}}(\tau) \\
&\quad - a^\dagger_{\mathbf{j}}(\tau)a^\dagger_{\mathbf{k}}(\tau)a_{\mathbf{j-q}}(\tau)a_{\mathbf{k+q}}(\tau)] \\
&\quad \bigotimes_{\substack{\mathbf{i}\neq\mathbf{j},\mathbf{k},\\ \mathbf{j-q},\\ \mathbf{k+q}}} \varrho^{\mathrm{eq}}_{\mathbf{i}}(a^\dagger_{\mathbf{x+y}} a^\dagger_{\mathbf{j-y}} a_{\mathbf{x}} a_{\mathbf{j}} - a^\dagger_{\mathbf{j}} a^\dagger_{\mathbf{x}} a_{\mathbf{j-y}} a_{\mathbf{x+y}})\} \\
&= -2\sum_{\mathbf{k},\mathbf{q}} |V(\mathbf{q})|^2 (g_{\mathbf{j}} g_{\mathbf{j-q}} g_{\mathbf{k+q}} f_{\mathbf{k}} + f_{\mathbf{j}} f_{\mathbf{j-q}} f_{\mathbf{k+q}} g_{\mathbf{k}}) \\
&\quad \times \cos[(\omega_{\mathbf{k+q}} + \omega_{\mathbf{j-q}} - \omega_{\mathbf{k}} - \omega_{\mathbf{j}})\tau],
\end{aligned} \tag{A11}$$

from this follows Eq. (16).

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[1] J. J. Quinn and R. A. Ferrell, Phys. Rev. **112**, 812 (1958).
[2] J. J. Quinn, Phys. Rev. **126**, 1453 (1962).
[3] L. P. Kadanoff and G. Baym, *Quantum Statistical Mechanics* (Benjamin, New York, 1962).
[4] M. Bauer, S. Pawlik, and M. Aeschlimann, Proc. SPIE **3272**, 201 (1998).
[5] M. Bauer and M. Aeschlimann, J. Electron Spectrosc. Relat. Phenom. **124**, 225 (2002).
[6] V. P. Zhukov, O. Andreyev, D. Hoffman, M. Bauer, M. Aeschlimann, E. V. Chulkov, and P. M. Echenique, Phys. Rev. B **70**, 233106 (2004).
[7] I. Campillo, J. M. Pitarke, A. Rubio, E. Zarate, and P. M. Echenique, Phys. Rev. Lett. **83**, 2230 (1999).
[8] P. M. Echenique, J. M. Pitarke, E. V. Chulkov, and A. Rubio, Chem. Phys. **251**, 1 (2000).
[9] P. M. Echenique, R. Berndt, E. V. Chulkov, T. Fauster, A. Goldmann, and U. Höfer, Surf. Sci. Rep. **52**, 219 (2004).
[10] J. Jäckle, *Einführung in die Transporttheorie* (Vieweg, Braunschweig, 1978).
[11] D. Pines and P. Nozieres, *The Theory of Quantum Liquids, Vol. I:*

Normal Fermi Liquids (Addison-Wesley, New York, 1989.
[12] G. D. Mahan, *Many Particle Systems* (Plenum, New York, 1990).
[13] C. Kittel, *Quantum Theory of Solids* (Wiley, New York, 1963).
[14] V. P. Zhukov, E. V. Chulkov, P. M. Echenique, A. Marienfeld, M. Bauer, and M. Aeschlimann, Phys. Rev. B **76**, 193107 (2007).
[15] A. L. Fetter and J. D. Walecka, *Quantum Theory of Many-Particle Systems* (McGraw-Hill, New York, 1971).
[16] D. A. Kirzhnits, *Field Theoretical Methods in Many-Body Systems* (Pergamon, New York, 1967).
[17] G. E. Brown, *Many-Body Problems* (North-Holland, Amsterdam, 1972).
[18] L. Hedin, Phys. Rev. **139**, A796 (1965).
[19] L. Hedin and S. Lundqvist, in *Solid State Physics: Advances in Research and Applications*, edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1969), Vol. 23, p. 1.
[20] E. V. Chulkov, A. G. Borisov, J.-P. Gauyacq, D. Sànchez-Portal, V. M. Silkin, V. P. Zhukov, and P. M. Echenique, Chem. Rev. (Washington, D.C.) **106**, 4160 (2006).
[21] V. P. Zhukov and E. V. Chulkov, J. Phys.: Condens. Matter **14**, 1937 (2002).
[22] C. D. Spataru, M. A. Cazalilla, A. Rubio, L. X. Benedict, P. M. Echenique, and S. G. Louie, Phys. Rev. Lett. **87**, 246405 (2001).
[23] W.-D. Schöne, R. Keyling, M. Bandić, and W. Ekardt, Phys. Rev. B **60**, 8616 (1999).
[24] M. Rohlfing and S. G. Louie, Phys. Rev. B **62**, 4927 (2000).
[25] Y. Ma and M. Rohlfing, Phys. Rev. B **75**, 205114 (2007).
[26] A. Marini, R. Del Sole, A. Rubio, and G. Onida, Phys. Rev. B **66**, 161104(R) (2002).
[27] I. A. Nechaev, I. Yu. Sklyadneva, V. M. Silkin, P. M. Echenique, and E. V. Chulkov, Phys. Rev. B **78**, 085113 (2008).
[28] S. Nakajima, Prog. Theor. Phys. **20**, 948 (1958).
[29] R. Zwanzig, J. Chem. Phys. **33**, 1338 (1960).
[30] H. Grabert, *Projection Operator Techniques in Nonequilibrium Statistical Mechanics* (Springer, Berlin, Heidelberg, 1982).
[31] P. Fulde, *Electron Correlations in Molecules and Solids* (Springer, Berlin, New York, 1993).
[32] J. Rau and B. Müller, Phys. Rep. **272**, 1 (1996).
[33] K. Burnett, J. Cooper, R. J. Ballagh, and E. W. Smith, Phys. Rev. A **22**, 2005 (1980).
[34] P. Neu and A. Würger, Z. Phys. B: Condens. Matter **95**, 385 (1994).
[35] H. P. Breuer and F. Petruccione, *The Theory of Open Quantum Systems* (Oxford University Press, Oxford, 2002).
[36] C. Bartsch, R. Steinigeweg, and J. Gemmer, Phys. Rev. E **77**, 011119 (2008).