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DYNAMICAL POLARIZABILITY OF ATOMS †

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ABSTRACT

The frequency-dependent polarizability of a closed-shell atom is considered in an RPA type approximation. This is usually done using many-body perturbation theory but can also be recast into the form of equations for the density oscillations as previously shown by the authors. The latter approach is known to lead to a non-hermitian problem because of the structure of the interaction kernel. This note shows that this is also true if using the reaction matrix method. The main result is to derive the expression for the polarizability function taking into account the non-hermitian nature of the problem.

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In recent years, considerable progress has been made in the application of random phase approximation (RPA) also including exchange (RPAE) and other many-body theories employing diagrammatic techniques and other methods to atoms with impressive success in explaining a number of experimentally observed genuine many-body effects particularly for heavy atoms. The subject has expanded now to a rather vast literature, and we refer the interested reader to Ref.1 for an extensive summary of the recent status of the field. The methods of calculation are usually based on variables which are discrete as well as continuous in nature, characterizing respectively the bound and continuous or resonant electronic states of the atom; in other words, working in Hilbert space. Recently, we have considered the possibility of the collective excitation of heavy atoms starting from the RPA in real space.[2),3)] This approach can be useful when one is interested in understanding the dynamic exchange and correlation effects in terms of the size of an exchange and correlation hole in the atom. The purpose of this paper is two-fold. The first is to establish contact between the real space approach of the standard RPA and the conventional approach in Hilbert space to find the dynamic dipolar polarizability $\alpha(\omega)$ of an atom. The second aim is to derive in detail the correct expression for $\mathrm{Im}\,\alpha(\omega)$ in the so-called reaction matrix method. We stay within RPA, but the results can easily be generalized to approximations beyond RPA.

When an atom is placed in a weak time-dependent external potential, the induced density can be calculated within linear response theory as [4)] (throughout we use units where $\hbar = 1$, $e^2 = 1$ and $m = 1$)

$$\rho_{ind}(\underset{\sim}{x},t) = \int d\underset{\sim}{x}' \int_{-\infty}^{t} dt'\, H(\underset{\sim}{x},\underset{\sim}{x}',t-t')\, V_{ext}(\underset{\sim}{x}',t') \quad , \qquad (1)$$

where $H(x,x',t)$ is the density-density response function of the atom. It is convenient to Fourier transform (1) in the time variable, so that (1) reads

$$\rho_{ind}(\underset{\sim}{x},\omega) = \int d\underset{\sim}{x}'\, H(\underset{\sim}{x},\underset{\sim}{x}',\omega)\, V_{ext}(\underset{\sim}{x}',\omega) \quad , \qquad (2)$$

where ω is the Fourier frequency. The Fourier-transformed density-density response function characterizes the response of the atom at the particular frequency ω. It is fairly easy to show from (2) that if the atom is placed in a uniform time-dependent electric field, the induced charge has the

character of an oscillating dipole, with dipolar polarizability given by

$$\alpha(\omega) = -\int d\underset{\sim}{x}\, d\underset{\sim}{x}'\, z\, z'\, H(\underset{\sim}{x}, \underset{\sim}{x}', \omega) \quad . \tag{3}$$

The quantity $H(x,x',\omega)$ is still unknown and here one uses the techniques of many-body theory. If one can find another density-density response function $h(x,x',\omega)$ that responds to the total potential, i.e. the external potential plus the induced potential due to the induced charge, then Eq.(2) can be used to obtain

$$H(\underset{\sim}{x}, \underset{\sim}{x}', \omega) = h(\underset{\sim}{x}, \underset{\sim}{x}', \omega) + \int d\underset{\sim}{x}_1\, d\underset{\sim}{x}_2\, h(\underset{\sim}{x}, \underset{\sim}{x}_1, \omega)\, v(\underset{\sim}{x}_1 - \underset{\sim}{x}_2)\, H(\underset{\sim}{x}_2, \underset{\sim}{x}', \omega), \tag{4}$$

where $v(\underset{\sim}{x}_1 - \underset{\sim}{x}_2) = |\underset{\sim}{x}_1 - \underset{\sim}{x}_2|^{-1}$, the Coulomb potential.

One has transferred the problem now to finding a suitable h. To lowest order approximation, h is given by the formula

$$h(\underset{\sim}{x}, \underset{\sim}{x}', \omega) = \sum_n \left[\frac{\varphi_n(\underset{\sim}{x})\, \varphi_n(\underset{\sim}{x}')}{\omega - \omega_n} - \text{n.f.} \right] \quad , \tag{5}$$

where it is understood that ω has a small positive imaginary part, i.e. $\omega \equiv \omega + i\epsilon$ (because we are dealing with retarded response that can give rise to real absorption); here n.f. stands for a term with ω_n replaced by $-\omega_n$ in the first term. In the so-called RPA, one has an excitation of frequency ω_n as a particle and hole pair excitation with corresponding wave functions $\psi_p(\underset{\sim}{x})$ and $\psi_h(\underset{\sim}{x})$ respectively and energies ϵ_p and ϵ_h respectively, so that $\omega_n = \epsilon_p - \epsilon_h$ and the wave function of the pair $\varphi_n(\underset{\sim}{x}) = \psi_p^*(\underset{\sim}{x})\, \psi_h(\underset{\sim}{x})$. In other words, the RPA expression for $h(x,x',\omega)$ is the so-called response function for non-interacting systems. One can go beyond RPA by suitably modifying the one-electron states $\psi(\underset{\sim}{x})$ and one-electron energies, which would then also require an appropriate modification of Eq.(4) still maintaining the essential structure. With the RPA form for h in Eq.(4), one can solve for H either by iteration or by the standard matrix inversion method. From the iteration method, it becomes transparent that RPA violates the Pauli principle. This is equivalent to neglecting the size of the exchange hole. In solids, RPA often work well, but there the size of the hole is small compared with the size of the system itself. In contrast, in

atoms, the size of the exchange-correlation hole can be comparable to the size of the atom itself, and violation of Pauli principle may cause a serious error. However, the number of electrons being small in atoms, it is possible, at least in principle, to correct Eq.(4) suitably, but here we shall ignore such questions. Finally, we note that Eqs.(1), (4) and (5) provided our starting point for obtaining a collective density oscillation in heavy atoms.[2),3)]

We now proceed to establish the connection between the above real space approach with the Hilbert space approach by defining the dipole moment of the n^{th} particle-hole pair as

$$p_n = \int d\underset{\sim}{x}\, z\, \varphi_n(\underset{\sim}{x}) \quad . \tag{6}$$

It is always possible to work with real one-particle wave functions, so that $\varphi_n(\underset{\sim}{x})$ is real whence p_n is also real, and henceforth we shall assume this to be the case. If now one substitutes Eq.(5) in Eq.(4) and uses the resulting equation in the expression (3), then one obtains

$$\alpha(\omega) = \sum_n \frac{2\omega_n p_n}{\omega_n^2 - \omega^2} (p_n - X_n) \quad , \tag{7}$$

where

$$X_n = -\int d\underset{\sim}{x}_1\, d\underset{\sim}{x}_2\, v(\underset{\sim}{x}_1 - \underset{\sim}{x}_2)\, \varphi_n(\underset{\sim}{x}_1) \int d\underset{\sim}{x}'\, z'\, H(\underset{\sim}{x}_2, \underset{\sim}{x}', \omega) \quad . \tag{8}$$

From the definition of X_n and Eq.(4) it follows that X_n is the solution of the integral equation

$$X_n = \sum_{n'} \frac{2\omega_{n'} p_{n'}}{\omega_{n'}^2 - \omega^2} V_{n'n} - \sum_{n'} \frac{2\omega_{n'}}{\omega_{n'}^2 - \omega^2} V_{n'n} X_{n'} \quad , \tag{9}$$

where $V_{nn'}$ is the usual Coulomb integral defined as

$$V_{nn'} = \int d\underset{\sim}{x}_1\, d\underset{\sim}{x}_2\, \varphi_n(\underset{\sim}{x}_1)\, v(\underset{\sim}{x}_1 - \underset{\sim}{x}_2)\, \varphi_{n'}(\underset{\sim}{x}_2) \equiv V_{n'n} \quad . \tag{10}$$

In the case of RPA with exchange (RPAE), one considers a second term added to (10) corresponding to the exchange process of two particle-hole pairs scattering through the Coulomb potential. Note that $2\omega_n p_n = f_n$, the so-called oscillator strength, and if we define $\gamma^2_{nm} = V_{nm}/p_n p_m$, then Eq.(9) takes a simple form which can be iterated to form a series representation for X_n and when the result is substituted in the expression (7) for $\alpha(\omega)$, one gets

$$\alpha(\omega) = \sum_n \frac{f_n}{\omega_n^2-\omega^2} - \sum_{nn'} \frac{f_n}{\omega_n^2-\omega^2}\,\gamma^2_{nn'}\,\frac{f_{n'}}{\omega_{n'}^2-\omega^2} + \cdots \quad . \tag{11}$$

This has the standard diagrammatic representation of RPA bubble series (see Fig.1) if one recognizes that a bubble represents the term $(\omega-\omega_n)^{-1}$ (i.e. a particular particle-hole pair excitation; a corresponding term for negative ω_n combines with it to produce the term $(\omega^2-\omega_n^2)^{-1}$ in Eq.(11)) and the line joining a pair of bubbles represents the factor $\gamma^2_{nn'}$ (i.e. the interaction between a pair of particle-hole excitations corresponding to frequencies ω_n and $\omega_{n'}$). Eq.(11) forms the basis of most of the many-body calculations for atoms.

We now turn to the problem of recasting the expression for Im $\alpha(\omega)$ in the reaction matrix method. We concentrate on the case of $\omega > 0$, and make use of the fact that $\omega \equiv \omega + i\epsilon$ implies that

$$(\omega_n^2-\omega^2)^{-1} = P\,(\omega_n^2-\omega^2)^{-1} + \frac{i\pi}{2\omega}\,\delta(\omega-\omega_n) \quad , \tag{12}$$

where P corresponds to taking only the principal value. The real and imaginary parts of Eq.(9) can be separated by using (12) to obtain

$$X'_n = \sum_{n'} \frac{V_{nn'}\,2\omega_{n'}p_{n'}}{\omega_{n'}^2-\omega^2} - \sum_{n'} \frac{V_{nn'}\,2\omega_{n'}}{\omega_{n'}^2-\omega^2}\,X'_{n'} + \pi\,V_{n\omega}\,X''_\omega \quad , \tag{13a}$$

and

$$X''_n = \pi\,V_{n\omega}\,(p_\omega - X'_\omega) - \sum_{n'} \frac{V_{nn'}\,2\omega_{n'}}{\omega_{n'}^2-\omega^2}\,X''_{n'} \quad , \tag{13b}$$

where X'_n and X''_n correspond respectively to the real and imaginary parts of X_n; $X'_\omega = \sum_n X'_n \delta(\omega-\omega_n)$ etc., and Σ means a principal value sum.

From Eq.(7) it then follows that

$$\alpha(\omega) = \sum_n 2\omega_n p_n \left[P\,(\omega_n^2-\omega^2)^{-1} + \frac{i\pi}{2\omega}\delta(\omega-\omega_n)\right]\left[(p_n-X'_n) - iX''_n\right] ,$$

so that

$$\mathrm{Im}\,\alpha(\omega) = \sum_n 2\omega_n p_n \left[\frac{\pi}{2\omega}\,\delta(\omega-\omega_n)(p_n-X'_n) - P\,(\omega_n^2-\omega^2)^{-1}\,X''_n\right] ,$$

$$= \pi\,p_\omega\,(p_\omega - X'_\omega) - \sum \frac{2\omega_n p_n}{\omega_n^2-\omega^2}\,X''_n \quad . \tag{14}$$

To solve Eqs.(13) we now introduce a real matrix K defined as

$$K_{nn'} = V_{nn'}\,P\,\frac{2\omega_{n'}}{\omega_{n'}^2-\omega^2} \quad . \tag{15}$$

Then Eq.(13b) reads

$$X''_n = \pi\,(p_\omega - X'_\omega)\,V_{n\omega} - \sum_{n'} K_{nn'}\,X''_{n'} \quad ,$$

$$= \sum_{n'} (1+K)^{-1}_{nn'}\,\pi\,(p_\omega - X'_\omega)\,V_{n'\omega} \quad ,$$

$$= \pi\,(p_\omega - X'_\omega)\,V_{n\omega}(\omega) \quad , \tag{16}$$

where

$$V_{nn'}(\omega) = \sum_{n''} (1+K)^{-1}_{nn''}\,V_{n''n'} \quad , \tag{17}$$

which is assumed to exist.

From Eqs.(15) and (17) it follows that

$$V_{nn'}(\omega) = V_{nn'} - \sum_{n''} V_{nn''}\,\frac{2\omega_{n''}}{\omega_{n''}^2-\omega^2}\,V_{n''n'}(\omega) \quad . \tag{18}$$

Putting expression (16) into (13a), we then obtain

$$X'_n = \sum_{n'} \frac{V_{nn'}\, 2\omega_{n'} P_{n'}}{\omega_{n'}^2 - \omega^2} + \pi^2 V_{n\omega}(P_\omega - X'_\omega) V_{\omega\omega}(\omega) - \sum_{n'} K_{nn'} X'_{n'} ,$$

$$= \sum_{n'n''} (1+K)^{-1}_{n\,n'} \frac{V_{n''n'}\, 2\omega_{n'} P_{n'}}{\omega_{n'}^2 - \omega^2} + \pi^2 V_{\omega\omega}(\omega)(P_\omega - X'_\omega) \sum_{n'} (1+K)^{-1}_{nn'} V_{n'\omega} ,$$

$$= \sum_{n'} V_{nn'}(\omega) \frac{2\omega_{n'} P_{n'}}{\omega_{n'}^2 - \omega^2} + \pi^2 V_{\omega\omega}(\omega)(P_\omega - X'_\omega) V_{n\omega}(\omega) . \qquad (19)$$

From Eq.(19) we can calculate X'_ω as follows:

$$X'_\omega = P_\omega - \frac{1}{1+\pi^2 V_{\omega\omega}^2(\omega)} \left[P_\omega - \sum_{n''} \frac{V_{\omega n''}(\omega)\, 2\omega_{n''} P_{n''}}{\omega_{n''}^2 - \omega^2} \right] . \qquad (20)$$

Putting (20) back into (16) and (19), we get

$$X''_n = \frac{\pi V_{n\omega}(\omega)}{1+\pi^2 V_{\omega\omega}^2(\omega)} \left[P_\omega - \sum_{n'} \frac{V_{\omega n'}(\omega)\, 2\omega_{n'} P_{n'}}{\omega_{n'}^2 - \omega^2} \right] , \qquad (21)$$

and

$$X'_n = \sum_{n'} V_{nn'}(\omega) \frac{2\omega_{n'} P_{n'}}{\omega_{n'}^2 - \omega^2} + \frac{\pi^2 V_{\omega\omega}(\omega) V_{n\omega}(\omega)}{1+\pi^2 V_{\omega\omega}^2(\omega)} \left[P_\omega - \sum_{n'} \frac{V_{\omega n'}(\omega)\, 2\omega_{n'} P_{n'}}{\omega_{n'}^2 - \omega^2} \right] . \qquad (22)$$

Putting these results,(21) and (22), into expression (14), we then obtain, after some algebra,

$$\mathrm{Im}\,\alpha(\omega) = \frac{\pi}{1+\pi^2 V_{\omega\omega}^2(\omega)} \left[P_\omega - \sum_{n'} \frac{V_{\omega n'}(\omega)\, 2\omega_{n'} P_{n'}}{\omega_{n'}^2 - \omega^2} \right] \left[P_\omega - \sum_{n} \frac{2\omega_n P_n V_{n\omega}(\omega)}{\omega_n^2 - \omega^2} \right] . \qquad (23)$$

If we now define

$$P^\dagger_\omega = P_\omega - \sum_n \frac{2 V_{\omega n}(\omega) P_n \omega_n}{\omega_n^2 - \omega^2} , \qquad (24a)$$

and

$$P_\omega = P_\omega - \sum_n \frac{2 P_n \omega_n V_{n\omega}(\omega)}{\omega_n^2 - \omega^2} , \qquad (24b)$$

we can then write a compact expression for $\mathrm{Im}\,\alpha(\omega)$

$$\mathrm{Im}\,\alpha(\omega) = \frac{\pi P_\omega^2}{1+\pi^2 V_{\omega\omega}^2(\omega)} + \frac{\pi (P^\dagger_\omega - P_\omega) P_\omega}{1+\pi^2 V_{\omega\omega}^2(\omega)} . \qquad (25)$$

It is only the first term that has been used in several calculations.[5]

It may be noted that the second term vanishes if $P^\dagger_\omega$ and P_ω are identical; in other words, if $V_{\omega n}(\omega) \equiv V_{n\omega}(\omega)$, i.e. if $V_{nn'}(\omega)$ are symmetric in n and n' . This would have been the case if $V_{nn'}$ were separable (see Eq.(18)), which is approximately true for excitations of very high frequencies, but in general there is no reason for the second term to vanish.

We wish to point out that the many-body interactions do in general lead to a non-hermitian problem involving non-symmetric interaction kernels. This was probably recognized for the first time in the pioneering paper by Bloch [6], in which he studied the collective oscillations of an atom developing a time-dependent Thomas-Fermi scheme. In this approach, if one of the equations describes the oscillating density, the adjoint equation describes the corresponding electrostatic potential and the eigensolutions form a complete bi-orthogonal set. The same properties hold if the space formulation, equivalent to HPA, developed in Ref.4 is being used. Our result in Eqs.(18), (24) and (25) shows that that diagrammatic many-body perturbation theory indeed leads to the same mathematical structure and thus the non-hermitian nature of the problem leads to an additional contribution which does not seem to have been included in the previous literature.

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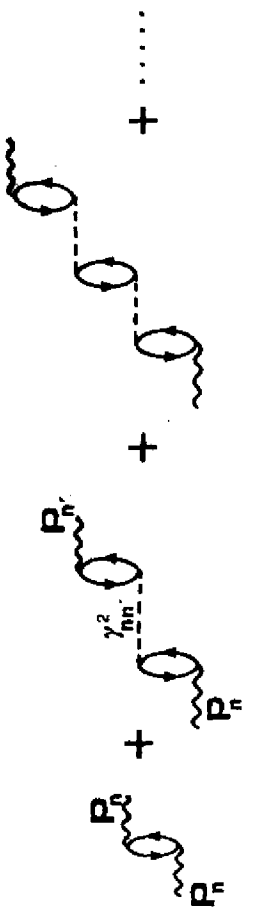
FIGURE CAPTION

Fig.1 Diagrammatic representation of Eq.(11) with only the positive frequency part shown. Each term will have to be summed over all possible excitations.



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