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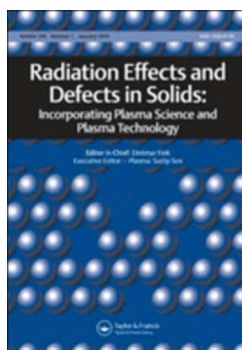
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Lindhard's polarization parameter and atomic sum rules in the local plasma approximation: A case for excited states

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Abstract

In this work, we analyze the effects of Lindhard polarization parameter, χ , on the sum rule, S_p , within the local-plasma approximation (LPA) as well as on the logarithmic sum rule $L_p = dS_p/dp$, in both cases for the system in an initial excited state. We show results for a hydrogenic atom with nuclear charge Z for the spherically symmetric ($l = 0$) states with $n = 1, 2$ and 3 with $p = -1, 0, 1$, and 2 . Our calculations are complemented with comparisons to the exact results for this system for the ground state. As a natural extension of our treatment, we report analytical results for helium-like systems in terms of a screened charge Z^* for the ground state. Our study shows that by increasing χ , the sum rule for $p < 0$ decreases while for $p > 0$ it increases, and the value $p = 0$ provides the normalization/closure relation which remains fixed to the number of electrons for the same initial state. When p is fixed, the value of S_p increases as the initial excited state increases for $p < 0$ and decreases for $p > 0$. Thus, when using a specific value of χ to adjust to a particular S_p sum rule value, the sum rule becomes unbalanced for other values of p . Our results show that the dipole oscillator strength distribution, within the LPA, provides good results for the sum rules for excited states.

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I. INTRODUCTION

Since the earliest days of quantum mechanics, dipole oscillator sum rules have played an important role for a quantum system. For example, the Thomas-Reiche-Kuhn (TRK) sum rule was used to describe the physics of electric-dipole interactions with atoms [1–3]. Other early uses of sum rules includes Bethe’s study of energy loss mechanisms for charged particles in matter [4] by the introduction of the mean excitation energy I_0 . Since then, sum rules have been used in many areas of physics, e.g. atomic [5, 6], molecular [7], solid state [8–10], nuclear [11] and particle physics [12].

The quantum mechanical dipole oscillator sum rule is defined as

$$S_p = \sum_{n \neq n_0} (E_n - E_{n_0})^p f_{nn_0} + \int E^p \frac{df}{dE} dE \quad (1)$$

and its logarithmic counterpart as

$$\begin{aligned} L_p &= \frac{dS_p}{dp} \\ &= \sum_{n \neq n_0} (E_n - E_{n_0})^p f_{nn_0} \ln(E_n - E_{n_0}) \\ &\quad + \int E^p \frac{df}{dE} \ln(E) dE \end{aligned} \quad (2)$$

where the sum is over the discrete spectrum and the integral over the continuum states of the system. Here f_{nn_0} and df/dE are the dipole oscillator strength and dipole oscillator strength spectrum for the discrete and continuum excitations from an initial state n_0 to a final state n , respectively. Even though p is an integer, we treat it formally as a continuous variable in Eqs. (1) and (2).

Sum rule relations contain a large amount of information about both the energy spectrum and energy eigenfunctions of the system in a compact form, particularly regarding closure relations. Such relations rely on the fact that the solutions of the system under study form a complete set of states. This constrains the application of such sum rules to systems where the bound and continuum states are known, a problem in many computational quantum physics applications.

When the whole excitation spectrum is known, closure relations allow the sum rule to be written in terms of initial state properties of the system. For example, Bethe and Jackiw

[13] show that

$$\begin{aligned}
 S_{-1} &= 2\langle n_0 | \mathbf{r}^2 | n_0 \rangle \\
 S_0 &= Z \\
 S_1 &= 2\langle n_0 | \mathbf{p}^2 | n_0 \rangle \\
 &= 4(E_{n_0} - \langle n_0 | V | n_0 \rangle) \\
 S_2 &= \langle n_0 | \nabla^2 V | n_0 \rangle \\
 &= 4\pi \langle n_0 | \rho(\mathbf{r}) | n_0 \rangle \\
 S_3 &= 2\langle n_0 | (\nabla V)^2 | n_0 \rangle
 \end{aligned} \tag{3}$$

and

$$L_0 = Z \ln I_0 \tag{4}$$

which are known as the quadrupole moment, TRK, momentum squared, force times momentum, force squared, and mean excitation energy sum rules [13], respectively. Here Z is the number of electrons in the system. Consequently, these sum rules allow us to test the excitation spectrum by comparing to the closure relations, e.g. by testing completeness of basis sets via the TRK sum rule [14].

However, when the system is treated within the local plasma approximation (LPA) such sum rule relations have a simpler form depending solely on the ground state density. In this approximation, one regards an atom or molecule as consisting of a sum of small volumes containing charge that independently responds to an external uniform electric field in the same way as would a free-electron gas. Applications of the LPA to the calculation of the mean excitation energy for stopping power in the Bethe theory are remarkably realistic, see for example Ref. [15].

In this work, we extend the use of the LPA to the study of sum rules when the system is initially in an excited state. We do so by means of the LPA dipole oscillator strength spectrum and analyze the effect of Lindhard's polarization parameter, χ , on the sum rule as a function of the sum rule order, p .

In Sec. II, we provide a review of the LPA sum rules. The analytical results obtained in this work for the hydrogenic benchmark system and a helium-like system are given in Sec. III, where we discuss our results. Finally in Sec. IV, we provide our conclusions. Atomic units are used throughout this work, except when physical units are specified.

II. THEORY

Lindhard and Scharff [16] used the idea of treating the system electronic charge distribution as a local plasma to determine photo-absorption properties of atoms. In an electron gas of uniform density ρ , the plasma oscillates with a frequency $\omega_p = \sqrt{4\pi\rho}$. Assuming that each volume element in the charge distribution contributes independently to the absorption strength, then each absorption frequency is equal to the amount of charge density at the given volume position, i.e $\omega = \omega(\mathbf{r})$.

Brandt and Lundqvist [17] and Brandt et al. [18] found, in the LPA, that the dipole oscillator strength spectrum can be written as

$$\frac{df^{LPA}}{dE} = \int \rho(\mathbf{r})\delta(E - \chi\omega_p(\mathbf{r}))d^3\mathbf{r}, \quad (5)$$

where $\rho(\mathbf{r})$ is the electronic density and χ is a parameter originally introduced by Lindhard and Scharff to account for polarization effects. It is customarily assumed that for most systems $\chi = \sqrt{2}$.

In this continuum representation of the electronic spectrum, Eqs. (1) and (2) together with Eq. (5) give us

$$S_p = (4\pi)^{p/2}\chi^p \int \rho(\mathbf{r})^{1+p/2}d^3\mathbf{r} \quad (6)$$

and

$$L_p = (4\pi)^{p/2}\chi^p \int \rho(\mathbf{r})^{1+p/2} \ln[\chi\omega(\mathbf{r})]d^3\mathbf{r} \quad (7)$$

which provide simple expressions for the sum rules in terms of the electronic density. We should note that these expressions are not valid for $p = -2$. Also, the case $p = 2$ completely reduces to the exact case in Eq. (4) via Poisson's equation $\nabla^2V = 4\pi\rho$ with an extra term χ^2 . It is precisely the appearance of this factor that pondered the question of its effect on the sum rules. Furthermore, it has been customary [15] to use the ground state electronic density of the system to determine the sum rules. Here, we will make use of the electronic density to incorporate the system initial state i.e. $\rho(\mathbf{r}) = \rho_{n_0}(\mathbf{r}) = |\psi_{n_0}(\mathbf{r})|^2$, where ψ_{n_0} is the initial state wave-function, as is assumed in Eqs. (1-2) and can be different from the ground state.

In the following section we provide results for the sum rules for initial states from $n_0 = 1$, 2, and 3 for a hydrogenic atom. We compare with the exact sum rules reported by Bethe and Jackiw [13] for the case $n_0 = 1$.

III. RESULTS

A. Hydrogen-like atoms

By using the exact hydrogenic wave-functions [13], we find that when the initial state is the ground state ($n_0 = 1$), the sum rule in Eq. (6) reduces to a simple analytical expression

$$S_p = \frac{2^{3+p} \chi^p Z^{3p/2}}{(2+p)^3}. \quad (8)$$

When $n_0 = 2$ and $l = 0$, we find that

$$\begin{aligned} S_p &= \frac{\chi^p Z^{3p/2}}{e^{2+p} (2+p)^5} 2^{2-p/2} \left(\pm \frac{(28+p(21+4p))}{(2+p)^{p-1}} \Gamma(2+p) \right. \\ &\quad \mp \frac{(5+p) \Gamma(3+p)}{(2+p)^p \Gamma(6+p)} \left[\pm 2e^{2+p} (2+p)^{6+p} \right. \\ &\quad \left. \left. + (28+21p+4p^2) \Gamma(5+p) - \Gamma(5+p, -2-p) \right] \right) \end{aligned} \quad (9)$$

where the \pm and \mp corresponds to even and odd p , respectively. For the case when the initial excited state is a $3s$, we find that

$$S_p = \chi^p Z^{3p/2} \begin{cases} 68.04 & , p = -1 \\ 1 & , p = 0 \\ 0.023199 & , p = 1 \\ 0.000964 & , p = 2 \\ 0.000109 & , p = 3 \end{cases} \quad (10)$$

As observed, the sum rule, S_p , has a dependence $(\chi Z^{3/2})^p$ times a factor that depends on p and the initial state n_0 for a hydrogenic-like atom. In Fig. 1, we show the polarization parameter dependence of the sum rule on p and χ when the hydrogenic system is in its ground state, $n_0 = 1$. We notice that increasing the value of χ , the sum rule decreases for $p < 0$ and increases for $p > 0$ for this initial state. The normalization/completeness condition, $p = 0$, does not depend on the value of χ , as it should. In the same figure, we show the exact results reported by Bethe and Jackiw [13] for the case of a H-like atom in the ground state $n_0 = 1$ (square symbols).

In Fig. 2, we show the sum rule as a function of the order p for several values of χ in the case of the initial excited state $2s$. We note that the effect of Lindhard's polarization parameter is less severe for $p > 0$ than when the sum rule is for the ground state for the same

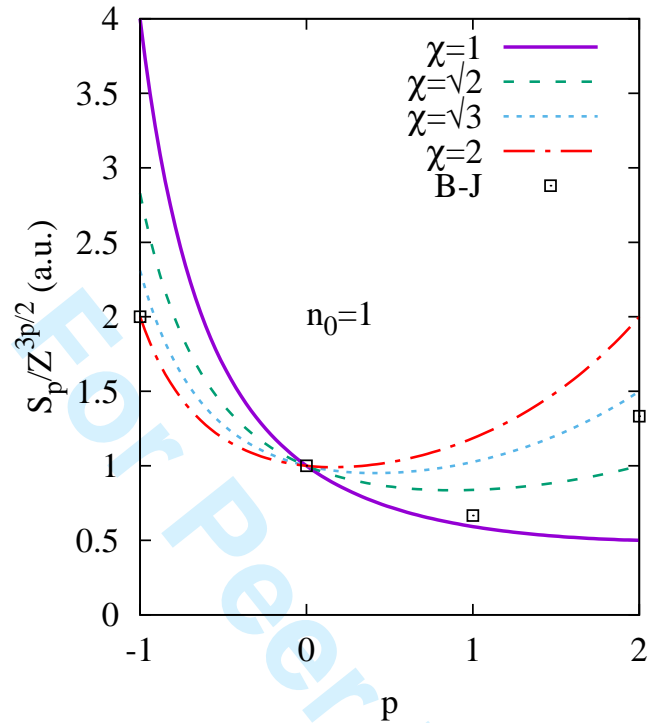


FIG. 1. Sum rule, S_p as a function of p for several values of Lindhard's polarization parameter, χ . The squared symbols are the exact values, as reported by Bethe and Jackiw [13].

value p . Also, the effect of χ on the sum rule is stronger for $p < 0$ than when $p > 0$ where the effect is minimal. A similar trend is observed for the case of excited state $3s$, as shown in Fig. 3. Thus, higher the excited initial state of the system, less sensitive is the LPA sum rule to the value of Lindhard's polarization parameter χ for $p > 0$. Also, we note that the sum rules decrease with increasing n_0 , except for S_0 which ensures normalization (number

TABLE I. Hydrogenic atom sum rule, $S_p/(\chi Z^{3/2})^p$, in a.u. as a function of p and the initial excited state n_0 . The values in parenthesis are the exact ground state results from Bethe and Jackiw [13].

$n_0 \backslash p$	-1	0	1	2
1	4	1	0.5926	$\frac{1}{2}$
	(2)	(1)	($\frac{2}{3}$)	($\frac{4}{3}$)
2	23.1555	1	0.0763	0.0098
3	68.0401	1	0.0232	0.0009

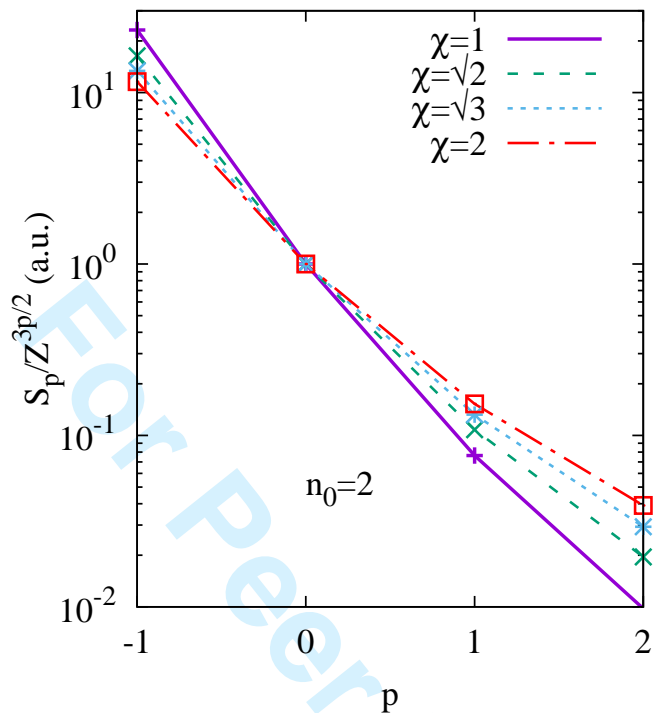


FIG. 2. Same as Fig. 1, but for excited state 2s.

of electrons in the system) for $p > 0$ and increases for $p < 0$. As it is known, the LPA is not that good for low density systems, as is a hydrogenic atom. However, we note that even in this case, we are in reasonable agreement with the exact results.

In Table I, we show the numerical evaluation of $S_p/(\chi Z^{3/2})^p$ for the case of a hydrogenic atom for $n_0 = 1, 2$, and 3. In the same tables, we compare with the exact results of Bethe and Jackiw [13] (in parenthesis) for $Z = 1$. In Table II, we show the value of L_p as obtained

TABLE II. Hydrogenic atomic sum rule L_p . The results in parenthesis are the mean excitation energy $I_0 = \exp(L_0/S_0)$ in units of eV.

$n_0 \backslash p$	-1	0	1	2
1	-9.2274	-0.8068	-0.1818	-0.0284
		(17.165)		
2	-86.1531	-2.7899	-0.1793	-0.0027
		(2.363)		
3	-317.3490	-	-	-

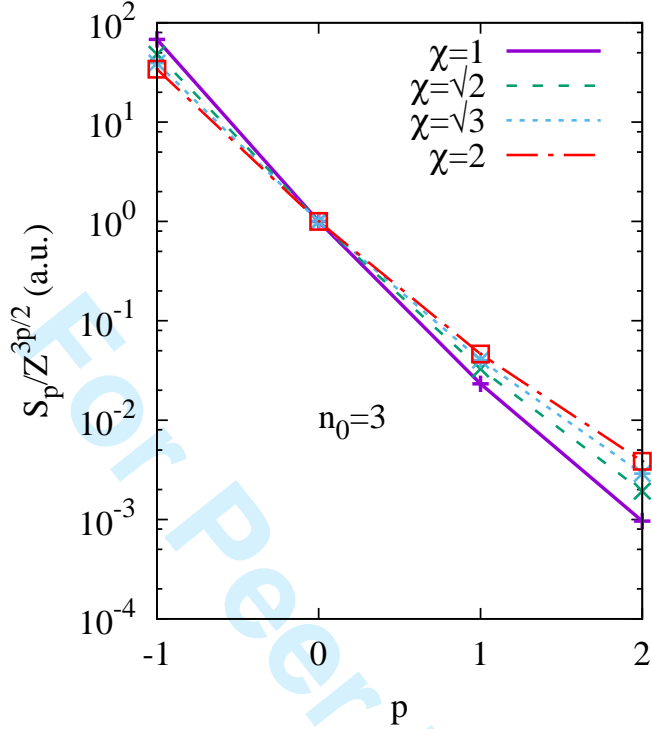


FIG. 3. Same as Fig. 1, but for excited state 3s.

by this approach for $\chi = 1$ and $Z = 1$. In parenthesis we show the corresponding values of $I_0 = \exp(L_0/S_0)$ in eV. The PSTAR result is $I_0 = 19.2$ eV as reported by [19] and our result is in good agreement within this approach. By increasing χ , our results agree much better to the exact case. In the same table we provide the mean excitation energy, I_0 for the atom in the initial 2s and 3s state.

B. He-like atoms

In the case of a helium-like atom, we use the simplest variational solution $\rho(r) = 2(Z^{*3}/\pi) \exp(-Z^*r)$ with a screening charge $Z^* = Z - 5/16$ in Eqs. (6) and (7), obtaining the following results

$$S_p = \frac{2^{4+\frac{3p}{2}} \chi^p Z^{*3p/2}}{(2+p)^3} \quad (11)$$

and

$$L_p = S_p \ln \left[\frac{\chi (2Z^*)^{3/2}}{e^{3/(2+p)}} \right]. \quad (12)$$

TABLE III. Helium-like atom sum rule S_p in a.u. as a function of p for the ground state for $Z = 2, 3$, and 4 with $\sqrt{\chi} = 2$.

$Z \backslash p$	-1	0	1	2
2	1.8247 2	5.1962	19.2217	
3	0.9079 2	10.4433	77.6436	
4	0.5649 2	16.7847	200.5650	

TABLE IV. The same as Table III but for L_p . The results in parenthesis are the mean excitation energy $I_0 = \exp(L_0/S_0)$ in units of eV.

$Z \backslash p$	-1	0	1	2
2	-1.5124	1.3423	6.0856	27.3172
		(53.216)		
3	-0.1187	2.7384	19.5208	164.5330
		(106.956)		
4	0.1941	3.6874	39.3387	520.2100
		(171.908)		

Note in this case that the S_p sum rule is a factor of 2 (the number of electrons) larger than the hydrogenic case, when $Z = Z^*$ for all values of p .

In Tables III and IV we provide numerical results for the sum rule S_p and L_p for helium-like atoms with $Z = 2, 3$, and 4 for $p = -1, 0, 1$, and 2 for the case of the initial state being the ground state. For this particular case, we use the value of $\sqrt{\chi} = 2$. In this case, the PSTAR value for I_0 is 41.8 eV [19], which our results is in good agreement with.

IV. CONCLUSIONS

We have used the LPA representation of the dipole oscillator strength distribution to calculate the sum rules S_p and $L_p = dS_p/dp$ when the system is found in an excited state. We provide results for hydrogen-like systems with nuclear charge Z . We also test the LPA approach for the sum rules for a helium-like system treated within a variational screened charge approach. We find that for higher excited states, the same sum rule decreases in value for $p > 0$ and increases for $p < 0$ and the effect of Lindhard's polarization parameter, χ

on the sum rules is to decrease S_p as χ increases for $p < 0$ and to decrease the value of the sum rule for $p > 0$ for fixed initial state n_0 . Thus, adjusting χ to a specific value will modify the sum rules for other values of p . We also show that both sum rules decrease as we move to higher and higher excited states for $p > 0$ and increases for $p < 0$.

We hope this work will motivate further research into more complicated atomic and molecular systems within this approach.

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