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Spatial-dispersion and relativistic effects in the optical sum rules

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Abstract. We describe a procedure to take into account the spatial dispersion of the optical excitations in the susceptibility sum rules. We show that this implies that relativistic corrections of the same order must be considered. The final result is a decrease of the total oscillator strength equal to the ratio of the average electron kinetic energy with mc^2. We propose experiments with synchrotron radiation sources on crystals of heavy elements to observe the described effect.

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1 Introduction

The optical sum rules have always attracted considerable attention because they are independent of any physical model and are strictly connected with general principles such as causality and inertia [1,2]. They are also of interest to experimentalists because they give information on the exhaustion of optical transitions and on the accuracy of all physical models; their validity has been experimentally verified in aluminum crystals by synchrotron radiation absorption and dispersion up to very high frequency [3,4].

A question which has not been sufficiently investigated is the role of spatial dispersion in the optical sum rules. It is generally assumed that the two variables ω and $\mathbf{k}$ on which the optical constants depend can be treated as independent quantities, but this is not the case for optical transverse modes because they must obey the dispersion equation [5]

$$\omega^2 = \frac{c^2 k^2}{\varepsilon(\mathbf{k},\omega)} \cdot \tag{1}$$

The problem of taking into account equation (1) in the Kramers-Kronig relations has been considered by Leontovich [6] and by Davydov [7], and a mathematical procedure has been suggested by Ginzburg and Meiman [8], who have first investigated the analytical properties of the optical functions $\varepsilon(\mathbf{k},\omega)$ and $n = \sqrt{\varepsilon}$ in the complex plane of the frequency variable.

Another point to be considered is the role played by relativistic corrections. The validity of the conventional sum rules is often limited to the dipole approximation, which is the lowest order in a relativistic expansion, and it corresponds, in crystals, to taking the limit $\mathbf{k} = 0$ in the dielectric function which appears in equation (1). It has been shown [9], for atoms, taking into account higher order relativistic corrections (magnetic dipoles, electric quadrupoles, etc.) in the independent-electron approximation, that the Thomas-Reiche-Kuhn (TRK) sum rule is modified in such a way that for any electron the oscillator strengths f_{0n} from state "0" to any state n do not sum to one, but we have:

$$\sum_n f_{0n} = 1 - \frac{\langle T \rangle_0}{mc^2} \tag{2}$$

where $\langle T \rangle_0$ denotes the average of the kinetic energy T on the ground state. A study of the relativistic corrections on the sum rule for the X-ray scattering factor has been recently carried out by Aucar, Oddershede and Sabin [10], who give a more accurate expression for the wavevector dependence of the sum rule, always considering photon momentum and frequency as independent variables.

In the present work we wish to study the sum rules of the optical response of solid materials taking into account both spatial dispersion and relativistic corrections. We will follow the suggestion of Ginzburg and Meiman [8] for the corrections due to spatial dispersion and will determine the constitutive relativistic equation for $\varepsilon(\mathbf{k},\omega)$.

In Section 2 we give the general theory of the relativistic response function with spatial dispersion. In Section 3 we discuss the analytic properties and the asymptotic

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behavior at high frequencies. In Section 4 we derive the sum rules. A discussion of the results is given in Section 5.

2 General theory

Following the approach of Ginzburg and Meiman [8], we define a new complex dielectric response function $\bar{\varepsilon}$ which depends on frequency only, by making use of equation (1) which allows to express $\mathbf{k}$ as a function of ω. We have, in an isotropic medium,

$$\bar{\varepsilon}(\omega) = \varepsilon\left(\hat{\mathbf{z}}\frac{\omega}{c}\sqrt{\bar{\varepsilon}(\omega)}, \omega\right), \tag{3}$$

where $\hat{\mathbf{z}}$ is the unitary vector that identifies the direction of propagation. Our first objective is to establish the existence of Kramers-Kronig relations for $\bar{\varepsilon}(\omega)$. This is equivalent to verifying the holomorphism of function (3) in the upper half of the complex ω plane, according to Titschmarsh's theorem [11]. Leontovich [6] demonstrated this property in the low density limit where it is appropriate to use the approximation $\bar{\varepsilon}(\omega) \simeq \varepsilon(\hat{\mathbf{z}}\omega/c, \omega)$. Ginzburg and Meiman [8], instead, make use of the expansion in powers of k_z, which, in isotropic and non-gyrotropic media reads

$$\bar{\varepsilon}(\omega) = \varepsilon(0,\omega) + \frac{k_z^2}{2}\frac{\partial^2\varepsilon}{\partial k_z^2}(\mathbf{k},\omega)|_{k=0} + O(k^4). \tag{4}$$

Where the linear term in k_z does not appear because it vanishes by time reversal. Higher than quadratic terms have been neglected due to the smallness of k in the optical modes. Using for k_z expressions (1) and (3), and substituting it into (4), we obtain, to order $1/c^2$,

$$\begin{aligned}\bar{\varepsilon}(\omega) &= \frac{\varepsilon(0,\omega)}{1 - \frac{\omega^2}{2c^2}\frac{\partial^2\varepsilon}{\partial k_z^2}(\mathbf{k},\omega)|_{k=0}} \\ &\simeq \varepsilon(0,\omega)\left[1 + \frac{\omega^2}{2c^2}\frac{\partial^2\varepsilon}{\partial k_z^2}(\mathbf{k},\omega)|_{k=0}\right]\end{aligned} \tag{5}$$

which coincides with the expression given by Ginzburg and Meiman [8]. This is an holomorphic function because so are $\varepsilon(0,\omega)$ and the second derivative appearing on the right hand side of equation (5). To obtain Kramers-Kronig relations for $\bar{\varepsilon}(\omega) - 1$ we need to show that this function goes to zero for $\omega \to \infty$ faster than ω^{-1}.

The asymptotic behavior of $\varepsilon(0,\omega)$ has been demonstrated in the non-relativistic approximation to be $1 - \omega_{\mathrm{p}}^2/\omega^2$ [1], where ω_{p} is the plasma frequency and this has been shown to be sufficient to obtain all the usual sum rules, since $\varepsilon(0,\omega)$ is holomorphic in the upper complex plane. We must now find the asymptotic behavior of expression (5) including spatial dispersion and relativistic corrections. This is necessary because spatial dispersion introduces terms of order $1/c^2$ and to be consistent also $\varepsilon(0,\omega)$ has to be calculated to order $1/c^2$. Consequently we can compute the susceptibility from the current density

$$\langle \mathbf{J}(\mathbf{k},\omega)\rangle = \frac{\omega^2}{c}\left[\frac{\varepsilon(\mathbf{k},\omega)}{4\pi}\right]\mathbf{A}(\mathbf{k},\omega). \tag{6}$$

To obtain the relativistic expression for the current density we must start from the Dirac definition of the current density of a given particle, in the presence of a potential $V(\mathbf{r})$ and of a radiation field expressed in terms of $\mathbf{A}(\mathbf{r},t)$. This is different from the approach of reference [9], because they introduce the relativistic corrections on the Hamiltonian operator only.

The relativistic definition of the current is [12]:

$$\begin{aligned}J_\alpha(\mathbf{r},t) = ec\sum_i [&\langle\phi|\sigma_\alpha\delta(\mathbf{r}-\mathbf{r}_i)|\chi\rangle \\ &+\langle\chi|\sigma_\alpha\delta(\mathbf{r}-\mathbf{r}_i)|\phi\rangle]\end{aligned} \tag{7}$$

where σ_α denotes the α Cartesian component of the Pauli matrices and ϕ and χ are the "large" and "small" components respectively of the four-spinors, which satisfy the equations:

$$(E' - V)|\phi\rangle = c\sigma_\alpha(p_\alpha - eA_\alpha/c)|\chi\rangle \tag{8}$$

$$(E' + 2mc^2 - V)|\chi\rangle = c\sigma_\alpha(p_\alpha - eA_\alpha/c)|\phi\rangle \tag{9}$$

where $E' + 2mc^2$ is the eigenvalue. Substituting (9) into equation (7) and expanding to order $1/c^2$ we obtain:

$$\begin{aligned}J_\alpha(\mathbf{r},t) = \frac{e}{2m}\sum_i \Bigg[&\langle\phi|\sigma_\alpha\delta(\mathbf{r}-\mathbf{r}_i)\left(1 - \frac{p^2}{4m^2c^2}\right) \\ &\times\sigma_\beta\left(p_\beta - \frac{eA_\beta}{c}\right)|\phi\rangle + \langle\phi|\sigma_\beta\left(p_\beta - \frac{eA_\beta}{c}\right) \\ &\times\left(1 - \frac{p^2}{4m^2c^2}\right)\delta(\mathbf{r}-\mathbf{r}_i)\sigma_\alpha|\phi\rangle\Bigg]\cdot\end{aligned} \tag{10}$$

We observe that this expression, when transformed in the expectation value of the quasi-relativistic wave function $\psi = N\phi$ [12], requires the normalization $N = 1 + p^2/8m^2c^2$. Developing expression (10), with normalization included, and following the procedure indicated in [12] we obtain

$$\begin{aligned}J_\alpha(\mathbf{r},t) = \frac{e}{2m}\sum_i \Bigg[&\langle\psi|\sigma_\alpha\left(1 + \frac{p^2}{8m^2c^2}\right)\delta(\mathbf{r}-\mathbf{r}_i) \\ &\times\left(1 - \frac{p^2}{4m^2c^2}\right)\sigma_\beta\left(p_\beta - \frac{eA_\beta}{c}\right)\left(1 + \frac{p^2}{8m^2c^2}\right)|\psi\rangle \\ &+\langle\psi|\left(1 + \frac{p^2}{8m^2c^2}\right)\sigma_\beta\left(p_\beta - \frac{eA_\beta}{c}\right)\left(1 - \frac{p^2}{4m^2c^2}\right) \\ &\times\delta(\mathbf{r}-\mathbf{r}_i)\sigma_\alpha\left(1 + \frac{p^2}{8m^2c^2}\right)|\psi\rangle\Bigg]\cdot\end{aligned} \tag{11}$$

Making use of the algebraic properties of the Pauli matrices and taking the spatial Fourier transform in the random phase approximation we obtain, from equation (11),

$$\mathbf{J}(\mathbf{k},t) = \mathbf{J}_0(\mathbf{k},t) - \frac{ne^2}{mc}\mathbf{A}(\mathbf{k},t) \tag{12}$$

where $n = N/\Omega$ is the electron density and $\mathbf{J}_0$ denotes the Fourier transform of the contribution in equation (11) which does not explicitly depend on the vector potential.

Since the last term of (12) is already linear in the perturbing field, we are left with evaluating the linear response of $\mathbf{J}_0$ to the field. This can be accomplished using time dependent perturbation theory, and the interaction Hamiltonian in the form [1]

$$H_{\mathrm{int}} = -\frac{\Omega}{c}\sum_{\mathbf{k}} \mathbf{J}_0(\mathbf{k})\cdot\mathbf{A}(-\mathbf{k},t). \tag{13}$$

The time evolution of $\mathbf{J}_0$, from linear response theory [13], is given by

$$\langle \mathbf{J}_0(\mathbf{k},t)\rangle = \int G(\mathbf{k},t')\mathbf{A}(\mathbf{k},t-t')\mathrm{d}t', \tag{14}$$

with the time-dependent response function given by

$$G(\mathbf{k},t) = -\frac{\Omega}{\mathrm{i}\hbar c}\left\langle [\mathbf{J}_0(-\mathbf{k},-t),\mathbf{J}_0(\mathbf{k},0)]\right\rangle_0 \theta(t), \tag{15}$$

θ being the Heaviside "step" function. Considering the time Fourier transform of equation (14) we obtain

$$\langle \mathbf{J}_0(\mathbf{k},\omega)\rangle = G(\mathbf{k},\omega)\mathbf{A}(\mathbf{k},\omega) \tag{16}$$

and making use of (12) we derive the expression for the total current

$$\langle \mathbf{J}(\mathbf{k},\omega)\rangle = G(\mathbf{k},\omega)\mathbf{A}(\mathbf{k},\omega) - \frac{ne^2}{mc}\mathbf{A}(\mathbf{k},\omega). \tag{17}$$

The above expression, recalling the definition (6), gives our desired susceptibility

$$\varepsilon(\mathbf{k},\omega) = 1 + \frac{1}{\omega^2}\left[4\pi c G(\mathbf{k},\omega) - \omega_{\mathrm{p}}^2\right] \tag{18}$$

where $\omega_{\mathrm{p}}^2 = 4\pi N e^2/\Omega m$.

3 Asymptotic behavior

The asymptotic behavior of $\varepsilon(\omega,0)$, with relativistic corrections, as required in (5), can be immediately obtained from (18), and is given by

$$\varepsilon(0,\omega) = 1 - \frac{\omega_{\mathrm{p}}^2}{\omega^2} + o(\omega^{-2}), \tag{19}$$

since $G(0,\omega)$ vanishes at infinity.

We can observe that relativistic corrections give vanishing contribution in the asymptotic behavior.

We must next consider spatial dispersion. To find the asymptotic behavior of $\bar{\varepsilon}(\omega)$ with spatial dispersion we need to compute the second derivative of expression (18) with respect to $\mathbf{k}$ and use it in equation (5). In this case we can use the nonrelativistic expression because it gives already a contribution of order $1/c^2$. In particular, since expression (18) only depends on $\mathbf{k}$ through $G(\mathbf{k},\omega)$, we can consider the asymptotic behavior in the frequency variable of the second derivative of G with respect to k, by making use of the time-dependent response function $G(\mathbf{k},t)$ and taking the limit for $t\to 0^+$. Since at infinite time the Green's function and all its time derivatives vanish, considering explicitly the lowest order terms in the expansion we obtain:

$$\frac{\partial^2 G}{\partial k_z^2}(\mathbf{k},\omega)|_{k=0} = \frac{1}{\mathrm{i}\omega}\left.\frac{\partial^2 G}{\partial k_z^2}(\mathbf{k},t)\right|_{\substack{k=0\\ t\to 0^+}} - \frac{1}{\omega^2}\left.\frac{\partial}{\partial t}\frac{\partial^2 G}{\partial k_z^2}(\mathbf{k},t)\right|_{\substack{k=0\\ t\to 0^+}} + o(\omega^{-2}). \tag{20}$$

We show in the Appendix that the first term to the right hand side of equation (20) vanishes and the second term gives a contribution which leads to the asymptotic behavior

$$\frac{\partial^2 \tilde{G}}{\partial k_z^2}(\mathbf{k},\omega)|_{k=0} = \frac{1}{\omega^2}\frac{2e^2 n}{m^3 c}\langle p^2\rangle_0 + o(\omega^{-2}). \tag{21}$$

We are now in a position to use the asymptotic behavior (21) into expression (5), by means of equation (18) and thus obtain the total asymptotic behavior of the spatial dependent dielectric function

$$\begin{aligned}\bar{\varepsilon}(\omega) &= \left[1-\frac{\omega_{\mathrm{p}}^2}{\omega^2}\right]\left[1+\frac{\omega_{\mathrm{p}}^2}{\omega^2}\frac{\langle p^2\rangle_0}{m^2c^2}\right] + O(\omega^{-3})\\ &= 1 - \frac{\omega_{\mathrm{p}}^2}{\omega^2}\left(1-\frac{\langle p^2\rangle_0}{m^2c^2}\right) + O(\omega^{-3}).\end{aligned} \tag{22}$$

4 Sum rules

The above desired asymptotic behavior (22), combined with the holomorphic properties of $\bar{\varepsilon}(\omega)$ on the upper half of the complex ω plane are sufficient to derive the Kramers-Kronig relations for $\bar{\varepsilon}(\omega)$, whose expressions are formally identical to the usual ones [1,2], and do not need do be reported here.

We then follow the usual procedure of comparing the asymptotic behavior (22) with the expression that originates from the Kramers-Kronig relations using the superconvergence theorem [1,14] and obtain the new sum rules,

with spatial dispersion. They are

$$\int_0^\infty \omega \mathrm{Im}\bar{\varepsilon}(\omega)\mathrm{d}\omega = \frac{\pi}{2}\omega_\mathrm{p}^2\left[1 - \frac{2\langle T\rangle_0}{3mc^2}\right] \tag{23}$$

and

$$\int_0^\infty \mathrm{Re}\left[\bar{\varepsilon}(\omega) - 1\right]\mathrm{d}\omega = 0 \tag{24}$$

where we have used the kinetic energy $\langle T\rangle_0$ averaged over all occupied states, which, in isotropic media, is related to $\langle p_x^2\rangle$ by $3\langle p_x^2\rangle_0 = 2m\langle T\rangle_0$. Equations (23, 24) give the desired sum rules with both spatial dispersion and relativistic corrections to order $1/c^2$.

We wish to remark that the spatial dispersion correction and the relativistic correction have to be considered to the same order, but only the dispersion contribution gives a non vanishing result to order $1/c^2$.

5 Discussion and conclusions

The new sum rules (23) and (24) derived in the previous section modify the TRK sum rule and the Altarelli-Dexter-Nussenzweig-Smith (ADNS) sum rule in two ways. First of all, the spatial dependence is included properly in the frequency dependence of the optical function through expression (5). Secondly, the relativistic effects are also included. As a result the nonrelativistic total sum is modified by the factor $[1 - \frac{2\langle T\rangle_0}{3mc^2}]$, which indicates a sum rule anomaly which increases with the atomic number of the elements of the crystal and with the binding energy of the electrons considered in the sum. By the virial theorem, in fact, $\langle T\rangle_0$ is the average binding energy of the electrons, and this in mc^2 units gives the magnitude of the anomaly.

Our result does not agree with the relativistic anomaly in the sum rule computed by Levinger *et al.* [9] for a Dirac electron, because they only include relativistic corrections in the Hamiltonian and not in the operators, as already noted in reference [10]. Our results also differ from the expression recently derived by Aucar, Oddershede and Sabin [10] for the relativistic corrections to the TRK sum rule, because they take the limit $k \to 0$ for the optical case and consequently do not consider the spatial dispersion. We think that the same correction to the sum rules must be valid in the atomic case as well as in all solid state cases when spatial dispersion is explicitly considered. The role of spatial dispersion is equivalent to that due to multipolar expansions, and differs in any case from the dipolar longitudinal relativistic correction given by Aucar *et al.* [10].

A verification of the above corrections to the TRK sum rule has never been considered in all experimental data now available, but may be relevant in the study of the susceptibility of heavy element crystals, because the new synchrotron radiation laboratories provide sources of continuous radiation up to frequencies in the far asymptotic range. A first verification of the optical sum rules was carried out by Shiles and Smith [3] on aluminum. In this case the relativistic correction to ω_p^2 is of order 10^{-3}, which could already be detected. With the synchrotron radiation now available in high energy sources like ESRF at Grenoble, one can easily reach the region of 100 keV, and explore the asymptotic behavior and the sum rule for heavier elements like Cu or Ag, where the correction, of order 10^{-1}, may be easily observed. A detailed analysis of the absorption spectrum of Si and Ge up to very high photon energy would complete the analysis of interband transition probabilities [15] to include the contributions from core states and the relevant corrections on the band structure and on the sum rule.

We wish to acknowledge Luciano Fonda for suggesting the relevance of relativistic corrections to sum rules. We are also indebted to Vladimir Agranovich for pointing out the importance of spatial dispersion effects.

Appendix

The aim of this appendix is to evaluate the derivatives, in expression (20), of the time-dependent response function $G(k_z, t)$, defined in (15).

The second derivative of G with respect to the wavevector can be written as

$$\begin{aligned}\frac{\partial^2 G}{\partial k_z^2}(k_z,t)\bigg|_{k_z=0} = -\frac{\Omega}{\mathrm{i}\hbar c}\Bigg\langle &\left[\frac{\partial^2 J_{0x}}{\partial k_z^2}(k_z,-t)\bigg|_{k_z=0}, J_{0x}(0,0)\right] \\ &- 2\left[\frac{\partial J_{0x}}{\partial k_z}(k_z,-t)\bigg|_{k_z=0}, \frac{\partial J_{0x}}{\partial k_z}(k_z,0)\bigg|_{k_z=0}\right] \\ &+ \left[J_{0x}(0,-t), \frac{\partial^2 J_{0x}}{\partial k_z^2}(k_z,0)\bigg|_{k_z=0}\right]\Bigg\rangle \end{aligned} \tag{A1}$$

where the current operator $J_{0x}(k_z, t)$ is defined, from (14) and (15) as

$$J_{0x}(k_z,t) = \frac{e}{m\Omega}\sum_i p_{ix}(t)\mathrm{e}^{-\mathrm{i}k_z z_i(t)}. \tag{A2}$$

We recall that we have to consider here only the nonrelativistic terms, because they already give a contribution of order $1/c^2$ in (5). The first derivative with respect to k_z that appears in (A1) is given by

$$\frac{\partial J_{0x}}{\partial k_z}(k_z,-t)\bigg|_{k_z=0} = -\frac{\mathrm{i}e}{m\Omega}\sum_i p_{ix}(-t)z_i(-t), \tag{A3}$$

while the second derivative is given by

$$\frac{\partial^2 J_{0x}}{\partial k_z^2}(k_z,-t)\bigg|_{k_z=0} = -\frac{e}{m\Omega}\sum_i p_{ix}(-t)z_i^2(-t). \tag{A4}$$

The $t \to 0^+$ limit of (A1), appearing in the r.h.s. of (20) is readily evaluated from (A1), using (A3) and (A4), and gives

$$\frac{\partial^2 G}{\partial k_z^2}(k_z,t)\bigg|_{k_z=0} = \frac{\Omega}{\mathrm{i}\hbar c}\frac{e^2}{m^2\Omega^2}\Bigg\langle \left[\sum_i p_{ix}z_i^2, \sum_i p_{ix}\right] - 2\left[\sum_i p_{ix}z_i, \sum_i p_{ix}z_i\right] + \left[\sum_i p_{ix}, \sum_i p_{ix}z_i^2\right]\Bigg\rangle = 0. \quad \text{(A5)}$$

This gives the general result that the first term to the r.h.s. of equation (20) vanishes.

The first derivative of (A1) with respect to time, which appears in the second term to the r.h.s. of (20), is evaluated from (A1) once the derivative of (A2, A3), and (A4) with respect to time are carried out. These are computed using the well-known expressions for the time derivative of the position and momentum operators

$$\frac{\mathrm{d}z_i}{\mathrm{d}t} = \frac{p_{ix}}{\mathrm{d}t}, \quad \frac{\mathrm{d}p_{ix}}{\mathrm{d}t} = -\frac{\partial V}{\partial x_i}\,. \quad \text{(A6)}$$

From (A2) we have

$$\frac{\partial J_{0x}}{\partial t}(0,-t)\bigg|_{k_z=0} = \frac{e}{m\Omega}\sum_i \frac{\partial V}{\partial x_i} \quad \text{(A7)}$$

and from (A3)

$$\frac{\partial}{\partial t}\frac{\partial J_{0x}}{\partial k_z}(k_z,-t)\bigg|_{k_z=0} = -\frac{\mathrm{i}e}{m\Omega}\sum_i\left(\frac{\partial V}{\partial x_i}z_i - p_{ix}\frac{p_{iz}}{m}\right) \quad \text{(A8)}$$

and finally, from (A4),

$$\frac{\partial}{\partial t}\frac{\partial^2 J_{0x}}{\partial k_z^2}(k_z,-t)\bigg|_{k_z=0} = -\frac{e}{m\Omega}\sum_i\left(\frac{\partial V}{\partial x_i}z_i^2 - \frac{p_{ix}}{m}(p_{iz}z_i + z_i p_{iz})\right). \quad \text{(A9)}$$

We are now in a position to compute the first derivative of (A1) with respect to time, which, making use of (A7, A8), and (A9), reads

$$\begin{aligned}\frac{\partial}{\partial t}\frac{\partial^2 G}{\partial k_z^2}(k_z,t)\bigg|_{k_z=0} =& \frac{\Omega}{\mathrm{i}\hbar c}\frac{e^2}{m^2\Omega^2}\Bigg\langle \left[\sum_i \frac{\partial V}{\partial x_i}z_i^2 + \frac{p_{ix}}{m}(p_{iz}z_i + z_i p_{iz}), \sum_i p_{ix}\right] \\ &+ 2\left[\sum_i \frac{\partial V}{\partial x_i}z_i + \frac{p_{ix}}{m}p_i z, \sum_i p_{ix}z_i\right] \\ &- \left[\sum_i \frac{\partial V}{\partial x_i}, \sum_i p_{ix}z_i^2\right]\Bigg\rangle \\ =& -\frac{2e^2}{m^3 c\Omega}\sum_i \langle p_{ix}^2\rangle_0.\end{aligned} \quad \text{(A10)}$$

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