Influence of a nonlocal potential on the induced current

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Outline

1. Motivations
2. How do we define the current
   - Definition
   - Simple cases
3. Coupling of nonlocal potentials to electromagnetic fields
   - Interaction hamiltonian
   - Induced current
   - Response function and sumrules
4. Summary

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Motivations

Problems

- Second harmonic generation: the calculation of the susceptibility shows some differences when comparing χ_{jjj} or $\chi_{\rho\rho\rho}$
- Sumrules

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Back to basics

Continuity equation - Charge conservation equation

$$div\,\mathbf{j}(\mathbf{r},t) + \frac{\partial \rho(\mathbf{r},t)}{\partial t} = 0$$

and in momentum space

$$\mathbf{k}.\mathbf{j}(\mathbf{k},t) + \frac{\partial \rho(\mathbf{k},t)}{\partial t} = 0 \qquad (1)$$

To fulfill the continuity equation, it is enough that Eq.(1) is fulfilled by current and density operators in the Heisenberg representation.

Heisenberg representation

$$\widehat{O}_H(t) = U^\dagger(t)\widehat{O}_S(t)U(t)$$

where $U(t)$ is the time evolution operator.

Expectation value of the density and current operators

Current

$$\mathbf{j}(\mathbf{r},t) = \frac{-i}{2}\sum_i < \psi(t)|\,[\mathbf{r},H(t)]\,\delta(\mathbf{r}-\mathbf{r}_i)+\delta(\mathbf{r}-\mathbf{r}_i)\,[\mathbf{r},H(t)]\,|\psi(t)>$$

In the independent-particle approximation

$$\mathbf{j}(\mathbf{r},t) = \frac{-i}{2}\sum_i f_i\,\{\psi_i^*(\mathbf{r},t)\,[\mathbf{r},H(t)]\,\psi_i(\mathbf{r},t) - [\mathbf{r},H(t)]\,\psi_i^*(\mathbf{r},t)\psi_i(\mathbf{r},t)\}$$

$$\rho(\mathbf{r},t) = \sum_i f_i|\psi_i(\mathbf{r},t)|^2$$

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The simplest case

Particle moving in a local potential

Hamiltonian:

$$H = \frac{1}{2}p^2 + V(\mathbf{r})$$

Commutator

$$[\mathbf{r}, H] = i\mathbf{p}$$

with $\mathbf{p} = -i\nabla$.
The current is defined by

$$\mathbf{j}(\mathbf{r}, t) = \frac{1}{2}\left\{\psi^*(\mathbf{r}, t)\mathbf{p}\psi(\mathbf{r}, t) + \mathbf{p}\psi^*(\mathbf{r}, t)\psi(\mathbf{r}, t)\right\}$$

Electromagnetic field

Free electron submitted to an electromagnetic field

Hamiltonian (minimal coupling convention) :

$$H = \frac{1}{2}\left[p - \frac{1}{c}\mathbf{A}(\mathbf{r},t)\right]^2 + \phi(\mathbf{r},t)$$

where ϕ and $\mathbf{A}$ are the scalar and vector potentials.
The commutator is

$$[\mathbf{r}, H] = i(\mathbf{p} - \frac{1}{c}\mathbf{A}(\mathbf{r},t))$$

Current

The current is defined by

$$\mathbf{j}(\mathbf{r},t) = \frac{1}{2}\Big\{\psi^*(\mathbf{p} - \frac{1}{c}\mathbf{A}(\mathbf{r},t))\psi + (\mathbf{p} - \frac{1}{c}\mathbf{A}(\mathbf{r},t))\psi^*\psi\Big\}$$

Note:the momentum $\mathbf{p}$ is replaced by $\mathbf{\Pi} = \mathbf{p} - \frac{1}{c}\mathbf{A}(\mathbf{r},t)$

Nonlocal potential

Particle moving in a nonlocal pseudopotential

Hamiltonian :

$$H = \frac{1}{2} p^2 + V_{nl}(\mathbf{r})$$

where V_{nl} is defined by $< \mathbf{r} | V_{nl} | \psi >= \int d\mathbf{r}' V_{nl}(\mathbf{r}, \mathbf{r}') \psi(\mathbf{r}')$
The commutator is

$$[\mathbf{r}, H] = i(\mathbf{p} - i\, [\mathbf{r}, V_{nl}])$$

The velocity operator is defined by $\mathbf{v} = \mathbf{p} - i\, [\mathbf{r}, V_{nl}]$ and we get

$$\mathbf{j}(\mathbf{r}, t) = \frac{1}{2} \left\{ \psi^* \mathbf{v} \psi + \mathbf{v} \psi^* \psi \right\}$$

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Question

Electromagnetic field

the momentum $\mathbf{p}$ is replaced by $\mathbf{\Pi} = \mathbf{p} - \frac{1}{c}\mathbf{A}(\mathbf{r}, t)$

$$\mathbf{j}(\mathbf{r}, t) = \frac{1}{2}\left\{\psi^*(\mathbf{p} - \frac{1}{c}\mathbf{A}(\mathbf{r}, t))\psi + (\mathbf{p} - \frac{1}{c}\mathbf{A}(\mathbf{r}, t))\psi^*\psi\right\}$$

Non-local potential

the momentum $\mathbf{p}$ is replaced by the velocity operator $\mathbf{v}$

$$\mathbf{j}(\mathbf{r}, t) = \frac{1}{2}\{\psi^*\mathbf{v}\psi + \mathbf{v}\psi^*\psi\}$$

Particle moving in a electromagnetic field and a nonlocal pseudopotential

Can we replace the momentum $\mathbf{p}$ by the operator $\mathbf{v} - \frac{1}{c}\mathbf{A}(\mathbf{r}, t)$?

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Minimal coupling prescription

Definition

The hamiltonian $H(\hat{\mathbf{r}}, \hat{\mathbf{p}})$ has to be changed, according to the substitution

$$\hat{\mathbf{p}} \longrightarrow \hat{\mathbf{\Pi}} = \hat{\mathbf{p}} - \frac{1}{c}\mathbf{A}(\hat{\mathbf{r}}, t)$$

For instance

$$\frac{1}{2}\hat{\mathbf{p}}^2 \longrightarrow \frac{1}{2}\hat{\mathbf{p}}^2 - \frac{1}{2c}\left\{\mathbf{A}(\hat{\mathbf{r}}, t)\hat{\mathbf{p}} + \hat{\mathbf{p}}\mathbf{A}(\hat{\mathbf{r}}, t)\right\} + \frac{1}{2c^2}A^2(\hat{\mathbf{r}}, t)$$

Minimal coupling prescription

Particle moving in a nonlocal potential

The nonlocal potential $V_{nl}(\mathbf{r},\mathbf{r}')$ can be considered as a local operator depending on **r** and **p**

$$< \mathbf{r}|V_{nl}|\Psi >= \int d\mathbf{r}' V_{nl}(\mathbf{r},\mathbf{r}')\Psi(\mathbf{r}') = V_{nl}(\mathbf{r},\mathbf{p})\Psi(\mathbf{r})$$

where $V_{nl}(\mathbf{r},\mathbf{p}) = \int d\mathbf{r}' V_{nl}(\mathbf{r},\mathbf{r}')e^{i(\mathbf{r}'-\mathbf{r})\mathbf{p}}$

The minimal coupling substitution has to be done also in V_{nl}

The coupled hamiltonian is $H_A = \frac{1}{2}\hat{\Pi}^2 + V(\hat{\mathbf{r}}) + \phi(\hat{\mathbf{r}},t) + V_{nl}^A$, where V_{nl}^A is defined by

$$< \mathbf{r}|V_{nl}^A|\mathbf{r}' >= V_{nl}(\mathbf{r},\mathbf{r}')e^{\frac{i}{c}\int_{\mathbf{r}'}^{\mathbf{r}} \mathbf{A}(\mathbf{x},t)d\mathbf{x}}$$

S. Ismail-Beigi, E.K. Chang and S. G. Louie, Phys. Rev. Lett. **87** 087402 (2001).

Perturbation theory

Interaction hamiltonian

We expand the hamiltonian in terms of **A** :

$$H_A = H_0 + H_1^{int} + H_2^{int} + \dots$$

To first order, a new term appears

$$H_1^{int} = -\frac{1}{2c}\{\mathbf{Ap} + \mathbf{pA}\} + \phi(\hat{\mathbf{r}}, t) + \frac{i}{c} V_{nl}(\mathbf{r}, \mathbf{r}') \int_{\mathbf{r}'}^{\mathbf{r}} \mathbf{A}(\mathbf{x}, t) d\mathbf{x}$$

In the long wavelength approximation ($\mathbf{A}(\mathbf{x}, t) = \mathbf{A}(t)$), one gets

$$H_1^{int} = -\frac{1}{c}\mathbf{Ap} + \phi(\hat{\mathbf{r}}, t) + \frac{i}{c}[\mathbf{A}(t)\mathbf{r}, V_{nl}] = -\frac{1}{c}\mathbf{Av} + \phi(\hat{\mathbf{r}}, t)$$

To second order : $H_2^{int} = -\frac{i}{2c^2}[\mathbf{Ar}, [\mathbf{Av}]]$

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Reminder

Current

Current:

$$\mathbf{j}(\mathbf{r},t) = \frac{-i}{2}\sum_i f_i \left\{\psi_i^*(\mathbf{r},t)\left[\mathbf{r},H(t)\right]\psi_i(\mathbf{r},t) - \left[\mathbf{r},H(t)\right]\psi_i^*(\mathbf{r},t)\psi_i(\mathbf{r},t)\right\}$$

coupling between a nonlocal potential and a electromagnetic field

Long wavelength limit

We have two terms in the hamiltonian :

$$H_0 = \frac{1}{2}p^2 + V_{nl}(\mathbf{r}) \qquad \text{and} \qquad H_1^{int} = -\frac{1}{c}\mathbf{A}\mathbf{v}$$

leading to the following (usual) current

$$\mathbf{j}^{(0)}(\mathbf{r},t) = \frac{1}{2}\left\{\psi^*(\mathbf{r},t)\mathbf{v}\psi_i(\mathbf{r}) + \mathbf{v}\psi_i^*(\mathbf{r})\psi_i(\mathbf{r},t)\right\}$$

and a new expression for the induced current

$$\mathbf{j}^{(1)}(\mathbf{r},t) = \frac{i}{2c}\left\{\psi^*(\mathbf{r},t)\left[\mathbf{r},\mathbf{A}.\mathbf{v}\right]\psi(\mathbf{r}) - \left[\mathbf{r},\mathbf{A}.\mathbf{v}\right]\psi^*(\mathbf{r})\psi(\mathbf{r},t)\right\}$$

Influence of the nonlocality

induced current for a local potential

$$\mathbf{j}^{(1)}(\mathbf{r},t) = -\frac{1}{c}\mathbf{A}(\mathbf{r},t))\rho(\mathbf{r},t)$$

induced current for a nonlocal potential

$$\mathbf{j}^{(1)}(\mathbf{r},t) = -\frac{1}{c}\mathbf{A}(\mathbf{r},t))\rho(\mathbf{r},t) + \frac{1}{2c}\left\{\psi^*(\mathbf{r},t)\left[\mathbf{r},\left[\mathbf{A}.\mathbf{r},V_{nl}\right]\right]\psi(\mathbf{r}) + \left[\mathbf{r},\left[\mathbf{A}.\mathbf{r},V_{nl}\right]\right]\psi^*(\mathbf{r})\psi(\mathbf{r},t)\right\}$$

$$\mathbf{p} \rightarrow \mathbf{v} - \frac{1}{c}\mathbf{A} + \frac{1}{c}\left[\mathbf{r},\left[\mathbf{A}.\mathbf{r},V_{nl}\right]\right]$$

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Long wavelength limit

In the limit $\mathbf{q} \rightarrow 0$

Density-density response function in the IPA:

$$\chi_{\rho\rho}(\mathbf{q},\mathbf{q},\omega) = \frac{2}{V}\sum_{nn'k}(f_{nk} - f_{n'k})\frac{< \phi_{nk}|\mathbf{qr}|\phi_{n'k} >< \phi_{n'k}|\mathbf{qr}|\phi_{nk} >}{(E_{nk} - E_{n'k} + \omega + i\eta)}$$

Current-current response function in the IPA:

$$\chi_{\mathbf{jj}}(\mathbf{q},\mathbf{q},\omega) = \frac{2}{V}\sum_{nn'\mathbf{k}}(f_{n\mathbf{k}} - f_{n'\mathbf{k}})\frac{< \phi_{n\mathbf{k}}|\mathbf{v}|\phi_{n'\mathbf{k}+\mathbf{q}} >< \phi_{n'\mathbf{k}+\mathbf{q}}|\mathbf{v}|\phi_{n\mathbf{k}} >}{(E_{n\mathbf{k}} - E_{n'\mathbf{k}+\mathbf{q}} + \omega)}$$

$$-\frac{2}{V}\sum_{n\mathbf{k}} f_{n\mathbf{k}} < \phi_{n\mathbf{k}}|\left[\mathbf{r},\left[\mathbf{r}, V_{nl}\right]\right]|\phi_{n\mathbf{k}} >$$

in terms of the Bloch functions.

Relation between the response functions

continuity equation

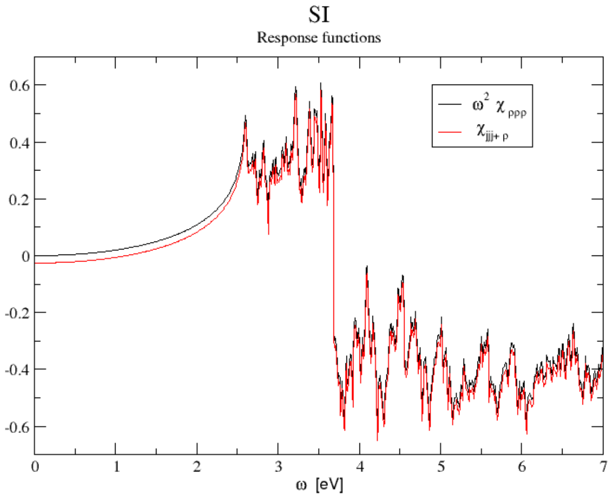
Taking into account

$$div\,\mathbf{j}(\mathbf{r},t) + \frac{\partial \rho(\mathbf{r},t)}{\partial t} = 0$$

one gets

$$\omega^2 \chi_{\rho\rho}(\mathbf{k},\mathbf{k}',\omega) = \mathbf{k}\left\{ < \rho(\mathbf{k} \hat{-} \mathbf{k}') > + \chi_{\mathbf{jj}}(\mathbf{k},\mathbf{k}',\omega) \right\} \mathbf{k}'$$

Relation between the response functions



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Conclusion

- In most cases $\chi_{\rho\rho}$ is used
- Even if χ_{jj} is calculated, the relation between the matrix elements of **v** and **r** is used to transform χ_{jj} into $\chi_{\rho\rho}$.
- The missing term is small
- The direct calculation of $\sum_{n\mathbf{k}} f_{n\mathbf{k}} < \phi_{n\mathbf{k}} | [\mathbf{q}.\mathbf{r}, [\mathbf{q}.\mathbf{r}, V_{nl}]] | \phi_{n\mathbf{k}} >$ turns out to be difficult.
  Stong anisotropy as a finction of **q** for cubic symmetry