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1986 J. Phys. A: Math. Gen. 19 3443

(http://iopscience.iop.org/0305-4470/19/16/039)

J. Phys. A: Math. Gen. 19 (1986) 3443-3447. Printed in Great Britain

On the gauge invariance of linear response theory

D Adu-Gyamfi†

International Centre for Theoretical Physics, Miramare, Trieste, Italy

Received 20 November 1985

Abstract. We demonstrate in a simple way, using a unitary transformation, the general gauge invariance of linear response theory.

1. Introduction

In studies of the response of a system of charged particles in an external electromagnetic field, the interaction between the particles and the field is usually expressed in the first instance as a linear functional of the electromagnetic potentials $\boldsymbol{A}(\boldsymbol{r}, t)$ and $\phi(\boldsymbol{r}, t)$, i.e.

$$H_1(t) = -\frac{1}{c}\int_V \mathrm{d}\boldsymbol{r}\, \boldsymbol{J}_0(\boldsymbol{r}) \cdot \boldsymbol{A}(\boldsymbol{r}, t) + \int_V \mathrm{d}\boldsymbol{r}\, d_0(\boldsymbol{r})\phi(\boldsymbol{r}, t). \tag{1.1}$$

In the above, $\boldsymbol{J}_0(\boldsymbol{r})$ and $d_0(\boldsymbol{r})$ are the current and charge densities, respectively, in the absence of external fields. The integration is over a volume V containing the system of particles. Since $H_1(t)$ depends on the potentials $\boldsymbol{A}(\boldsymbol{r}, t)$ and $\phi(\boldsymbol{r}, t)$, the use of the above interaction Hamiltonian entails having to establish gauge invariance in each specific case (Nakajima 1956, Kadanoff and Martin 1961, Siskens and Mazur 1972).

However, it has been established that by means of a canonical transformation on the non-relativistic Pauli Hamiltonian of a molecule in an external electromagnetic field, one can eliminate the potentials $\boldsymbol{A}(\boldsymbol{r}, t)$ and $\phi(\boldsymbol{r}, t)$ in favour of the electric and magnetic fields $\boldsymbol{E}(\boldsymbol{r}, t)$ and $\boldsymbol{B}(\boldsymbol{r}, t)$ apart from terms proportional to the total molecular charge. The fields and potentials are related in the usual manner by

$$\begin{aligned} \boldsymbol{E}(\boldsymbol{r}, t) &= -\frac{1}{c}\frac{\partial}{\partial t}\boldsymbol{A}(\boldsymbol{r}, t) - \nabla\phi(\boldsymbol{r}, t) \\ \boldsymbol{B}(\boldsymbol{r}, t) &= \nabla \wedge \boldsymbol{A}(\boldsymbol{r}, t). \end{aligned} \tag{1.2}$$

The canonical transformation which is unitary is defined by

$$\begin{aligned} U &= \exp\left(\frac{\mathrm{i}}{\hbar} g(t)\right) \\ g(t) &= -\frac{1}{c}\int_V \mathrm{d}\boldsymbol{r}\, \boldsymbol{P}(\boldsymbol{r}) \cdot \boldsymbol{A}(\boldsymbol{r}, t) \end{aligned} \tag{1.3}$$

where $\boldsymbol{P}(\boldsymbol{r})$ is the total electric polarisation density vector. The canonical transformation technique has, over the years, been considered by several workers including

† Permanent address: Department of Mathematics, University of Cross River State, Uyo, CRS Nigeria.

0305-4470/86/163443+05$02.50

Goeppert-Mayer (1931), Richards (1948), Lamb (1952), Power and Zienau (1959), Fiutak (1963), Atkins and Woolley (1970), Babiker *et al* (1973, 1974), Felderhof (1974), Felderhof and Adu-Gyamfi (1974) and Adu-Gyamfi (1981).

Generally, after the application of the unitary transformation, the initial non-relativistic Pauli Hamiltonian is reduced to multipole form and for a neutral molecule the potential terms are completely eliminated in favour of the electric and magnetic fields. Hence, this approach is particularly well suited for electromagnetic response theory and has recently been exploited to identify the 'electric' and 'magnetic' response terms for a neutral molecule (Adu-Gyamfi 1981). Nevertheless for ions some potential terms still survive and the issue of gauge invariance still remains relevant.

Barron and Gray (1973) demonstrated that a canonical transformation may not be necessary to transform the initial potential-dependent Hamiltonian to a field-dependent one, provided one chooses an appropriate gauge to start with. Following this, Woolley (1973, 1974) pointed out the equivalence of the gauge transformation approach and the canonical transformation formalism by choosing an initial gauge in which the corresponding generating function of the unitary transformation U vanishes, i.e.

$$g(t) = -\frac{1}{c}\int_V \mathrm{d}\boldsymbol{r}\, \boldsymbol{P}(\boldsymbol{r}) \cdot \boldsymbol{A}^*(\boldsymbol{r}, t) \equiv 0. \tag{1.4}$$

In effect one has the identity unitary transformation. This establishes a close relation between the gauge and canonical transformation approaches.

The demonstration of the general gauge invariance of linear response theory will be shown to follow from the invariance of linear response under a unitary transformation coupled with the fact that a classical gauge transformation is equivalent in the quantum mechanical formalism to an appropriate unitary transformation.

In § 2 we outline briefly the basic linear response theory of Kubo. This is shown, in § 3, to be invariant under a general unitary transformation. This in turn establishes the gauge invariance of linear response theory, through the equivalence of a classical gauge transformation with an appropriate quantum mechanical unitary transformation.

2. The basic linear response theory

In the conventional linear response theory (Kubo 1957), one considers a system which is slowly drawn out of its equilibrium state in such a way that the deviation from the equilibrium state is small. The perturbation responsible for this process is expressed by an interaction Hamiltonian $H_1(t)$ which allows one to find the change produced in the density matrix and, correspondingly, in the dynamical variables of the system. For perturbations which are electromagnetic in character, $H_1(t)$ is usually given by

$$H_1(t) = -\frac{1}{c}\int_V \mathrm{d}\boldsymbol{r}\, \boldsymbol{J}_0(\boldsymbol{r}) \cdot \boldsymbol{A}(\boldsymbol{r}, t) + \int_V \mathrm{d}\boldsymbol{r}\, d_0(\boldsymbol{r})\phi(\boldsymbol{r}, t)$$

in familiar notation.

The initial ensemble statistically representing the initial state of the system is specified by the equilibrium density matrix ρ_0 satisfying

$$\dot{\rho}_0 = \frac{1}{\mathrm{i}\hbar}[H_0, \rho_0] = 0 \tag{2.1}$$

where H_0 is the Hamiltonian in the unperturbed state. Under the external perturbation $H_1(t)$, the statistical distribution of the system is then described by the time-dependent density matrix $\rho(t)$ satisfying

$$\dot{\rho}(t)=\frac{1}{\mathrm{i}\hbar}[H_0+H_1(t),\rho(t)]$$

subject to

$$\rho(-\infty)=\rho_0=\exp(-\beta H_0)/\mathrm{Tr}(\exp(-\beta H_0)). \tag{2.2}$$

To first order in $H_1(t)$ we can express $\rho(t)$ as

$$\rho(t)=\rho_0+\rho_1(t)$$

where

$$\dot{\rho}_1(t)=\frac{1}{\mathrm{i}\hbar}[H_0,\rho_1(t)]+\frac{1}{\mathrm{i}\hbar}[H_1(t),\rho_0]. \tag{2.3}$$

This has the solution

$$\rho_1(t)=\frac{1}{\mathrm{i}\hbar}\int_{-\infty}^{t}\mathrm{d}t\exp\left(-\frac{\mathrm{i}}{\hbar}(t-\tau)H_0\right)[H_1(\tau),\rho_0]\exp\left(\frac{\mathrm{i}}{\hbar}(t-\tau)H_0\right). \tag{2.4}$$

The average of an observable O is determined according to

$$\langle O(t)\rangle=\mathrm{Tr}\{\rho(t)O(t)\}. \tag{2.5}$$

To first order in $H_1(t)$ we expand $O(t)$ as

$$O(t)=O_0+O_1(t) \tag{2.6}$$

where O_0 is the observable in the unperturbed state. Thus to first order in $H_1(t)$,

$$\begin{aligned}\langle O(t)\rangle_1&=\mathrm{Tr}\{\rho(t)O(t)\}\\&=\mathrm{Tr}\{\rho_0O_0\}+\mathrm{Tr}\{\rho_0O_1(t)\}+\mathrm{Tr}\{\rho_1(t)O_0\}\\&=\langle O_0\rangle_0+\langle O_1(t)\rangle_0+\frac{1}{\mathrm{i}\hbar}\int_{-\infty}^{t}\mathrm{d}\tau\langle[O_0(t-\tau),H_1(\tau)]\rangle_0.\end{aligned} \tag{2.7}$$

In the above, $\langle\ \rangle_0$ represents an equilibrium average. Without any loss of generality we let $\langle O_0\rangle_0$ vanish and define the linear response of O as

$$\langle O(t)\rangle_1=\langle O_1(t)\rangle_0+\frac{1}{\mathrm{i}\hbar}\int_{-\infty}^{t}\mathrm{d}\tau\langle[O_0(t-\tau),H_1(\tau)]\rangle_0. \tag{2.8}$$

In the case of electromagnetic perturbation, $H_1(t)$ is potential dependent, equation (1.1), and thus the linear response will be potential dependent. However, in appropriate cases it may be reduced to a linear relation in the fields $\boldsymbol{E}(\boldsymbol{r},t)$ and $\boldsymbol{B}(\boldsymbol{r},t)$ often after invoking various commutation relations and the continuity equation for charge and current densities (Siskens and Mazur 1972).

3. Unitary transformation and gauge invariance

As pointed out earlier, a classical gauge transformation

$$\boldsymbol{A}\to\boldsymbol{A}^*=\boldsymbol{A}+\nabla\chi \qquad \phi\to\phi^*=\phi-\frac{1}{c}\frac{\partial}{\partial t}\chi \tag{3.1}$$

is equivalent in the quantum mechanical description to a unitary transformation $U=\exp[\mathrm{i}g(t)/\hbar]$. The generating function $g(t)$ of the unitary transformation U is related to the generating function $\chi(\boldsymbol{r}, t)$ of the corresponding gauge transformation by

$$g(t)=\sum_{j=1}^{N}\frac{e_j}{c}\chi(\boldsymbol{r}_j, t) \tag{3.2}$$

where e_j, $\boldsymbol{r}_j$ represent the charge and position of the jth particle. Hence, we see that gauge invariance will follow simply from the invariance of the linear response under a unitary transformation U.

Under a unitary transformation $U=\exp[\mathrm{i}g(t)/\hbar]$ the Hamiltonian H, a linear operator O, wavefunction ψ and the density matrix ρ transform as follows:

$$\begin{aligned}
&H\to H''=UHU^{-1}-\partial g/\partial t\\
&O\to O'=UOU^{-1}\\
&\psi\to\psi'=U\psi\\
&\rho\to\rho'=U^{-1}\rho U.
\end{aligned}\tag{3.3}$$

Thus after the unitary transformation, we have to first order in $H_1(t)$

$$H''(t)=H_0+H_1''(t)$$

where

$$H_1''(t)=H_1(t)+\frac{\mathrm{i}}{\hbar}[g(t), H_0]-\frac{\partial g}{\partial t}$$

and

$$O'(t)=O_0+O_1'(t)$$

where

$$O_1'(t)=O_1(t)+\frac{\mathrm{i}}{\hbar}[g(t), O_0]. \tag{3.4}$$

Similarly the first-order correction to the equilibrium density matrix ρ_0 after the unitary transformation is given by

$$\rho_1'(t)=\frac{1}{\mathrm{i}\hbar}\int_{-\infty}^{t}\mathrm{d}\tau\exp\left(-\frac{\mathrm{i}}{\hbar}(t-\tau)H_0\right)[H_1''(\tau), \rho_0]\exp\left(\frac{\mathrm{i}}{\hbar}(t-\tau)H_0\right). \tag{3.5}$$

Hence, in the transformed framework, the linear response is given by

$$\begin{aligned}
\langle O'(t)\rangle_1' &= \mathrm{Tr}\{\rho'(t)O'(t)\}\\
&=\mathrm{Tr}\{\rho_0O_1'(t)\}+\mathrm{Tr}\{\rho_1'(t)O_0\}\\
&=\mathrm{Tr}\{\rho_0O_1(t)\}+\frac{\mathrm{i}}{\hbar}\mathrm{Tr}\{\rho_0[g(t), O_0]\}+\mathrm{Tr}\{\rho_1(t)O_0\}-\frac{\mathrm{i}}{\hbar}\mathrm{Tr}\{\rho_0[g(t), O_0]\}\\
&=\mathrm{Tr}\{\rho_0O_1(t)\}+\mathrm{Tr}\{\rho_1(t)O_0\}\\
&=\langle O(t)\rangle_1.
\end{aligned}\tag{3.6}$$

Therefore we have established the invariance of linear response under an arbitrary unitary transformation. This in turn establishes the gauge invariance of linear response theory. We note that the above proof is general and not restricted to any specific model.

Acknowledgments

The author would like to thank Professor Abdus Salam, the International Atomic Energy Agency and UNESCO for hospitality at the International Centre for Theoretical Physics, Trieste.

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