Annals of Physics 338 (2013) 260–277

Contents lists available at ScienceDirect

Annals of Physics

journal homepage: www.elsevier.com/locate/aop

Gauge-invariant Green function dynamics: A unified approach

Sylvia D. Swiecicki *, J.E. Sipe

Department of Physics and Institute for Optical Sciences, University of Toronto, 60 St. George Street, Toronto, Ontario, Canada M5S 1A7

HIGHLIGHTS

- Peierls phase for an arbitrary path in space–time established.
- Gauge-invariant Green functions and the Power–Zienau–Wooley transformation connected.
- Limitations on possible polarization and magnetization fields established.

ARTICLE INFO

Article history:
Received 3 August 2013
Accepted 18 September 2013
Available online 25 September 2013

Keywords:
Gauge-invariant Green function
Power–Zienau–Wooley transformation
Generalized Peierls transformation
Optical response

ABSTRACT

We present a gauge-invariant description of Green function dynamics introduced by means of a generalized Peierls phase involving an arbitrary differentiable path in space–time. Two other approaches to formulating a gauge-invariant description of systems, the Green function treatment of Levanda and Fleurov [M. Levanda, V. Fleurov, J. Phys.: Condens. Matter 6 (1994) 7889] and the usual multipolar expansion for an atom, are shown to arise as special cases of our formalism. We argue that the consideration of paths in the generalized Peierls phase that do not lead to introduction of an effective gauge-invariant Hamiltonian with polarization and magnetization fields may prove useful for the treatment of the response of materials with short electron correlation lengths.

1. Introduction

While at a fundamental level the interaction of the electromagnetic field with charges is described by the minimal coupling Hamiltonian, for applications in atomic and molecular physics it is usually more convenient to use the transformed Hamiltonian developed by Power, Zienau, and Wooley

* Corresponding author. Tel.: +1 416 823 1546.
E-mail addresses: sswiecic@physics.utoronto.ca (S.D. Swiecicki), sipe@physics.utoronto.ca (J.E. Sipe).

0003-4916/$ – see front matter
http://dx.doi.org/10.1016/j.aop.2013.09.014

(PZW) [1], and elaborated and clarified by Healy [2]. In the PZW transformation the interaction Hamiltonian is converted to one involving polarization and magnetization fields interacting directly with the electric and magnetic fields, which is thus gauge invariant. Often one then expands the polarization and magnetization fields in terms of electric and magnetic multipole moments, yielding an approximate Hamiltonian that takes into account the variations of the electric and magnetic fields over the atom or molecule to the order required. Even before such an expansion is undertaken, a "special point" must be chosen to effect the PZW transformation. For an atom this special point is often taken to be the position of the nucleus, idealized as fixed; more generally it can be taken to be the position of the center of mass of an atom or molecule [2].

Another strategy for constructing gauge-invariant quantum dynamical equations is the approach of Levanda and Fleurov (LF) [3,4]. They showed that by concentrating on the equations of motion for the Green functions, rather than modifying the Hamiltonian formulation of those equations, a gauge-invariant description of the dynamics is possible. Their approach employs a generalized Peierls phase that involves an integration along a straight line in space–time between the two Green function variables; see also [5] for a discussion and [6] for the extension to superconductors. The LF approach is attractive because it does not rely on a special point, as does the PZW transformation. Thus this kind of approach, or a related one, could provide a basis for gauge-invariant descriptions of the interaction of radiation with charges in extended systems, such as solids, where the use of a special point does not seem to make physical sense. Gauge-invariant descriptions would be useful because the straightforward application of the minimal coupling Hamiltonian often requires the identification of sum rules to eliminate apparent divergences, even in problems as simple as the linear and nonlinear optical response of clean, cold semiconductors, with electrons treated in the independent particle approximation [7–10].

These apparent divergences are associated with the difference between the mechanical momenta of the charges and their canonical momenta, described by a term involving the vector potential, and they seem to be avoided whenever a gauge-invariant description of the quantum dynamics can be constructed. There is a long tradition of work on special cases where this has been implemented. For situations where the response of the medium can be approximated as responding locally to the electric field, taken as uniform, an approach originally pioneered by Adams and Blount [11] can be applied. A quantity that plays the role of the matrix element of a dipole moment operator between Bloch states can be introduced, involving interband and intraband contributions [12]. The approach of the "modern theory of polarization" [13], where polarization and magnetization are associated with the appropriate moments of Wannier functions [14,15], or extensions necessary in materials with nonzero Chern numbers [16], can be seen to be closely related to this, for these moments can be written in terms of the effective matrix elements of Adams and Blount and their generalizations. But to date these programs are restricted to the assumption of uniform or nearly uniform applied fields.

So an exploration of strategies based on the LF approach to treat the response of solids to more arbitrarily varying electromagnetic fields seems promising. A natural first step would be to link that approach with the PZW transformation, which is to date probably the most successful gauge-invariant description of the response of charges to the electromagnetic field. Yet when both methods are applied to an atom, the equivalence can be seen only after much manipulation, even when the electromagnetic field varies little over the atom. The first goal of this paper is to clarify the link between the gauge-invariant descriptions of LF and PZW.

To do this we find it is necessary to extend the LF approach to treat a Peierls phase involving an arbitrary path in space–time connecting the two Green function variables. This is done in Section 2, where we construct a generalized LF approach (GLF). Then we can recover the LF and PZW results by the choice of particular paths. This is done in Sections 3 and 4.

Establishing those connections illustrates that there are two qualitatively different types of Peierls phase that can arise in a GLF approach. In the first type, which appears in recovering the PZW results, the generalized Peierls phase is given by the difference of the values that a single variable function takes at the two space–time points in the Green function. In the second type, which appears for example in the original LF approach, it cannot be written in that form. In Section 4 we show that the Peierls phase of the first type leads to the gauge-invariant Green functions that can be understood as constructed from transformed field operators, which satisfy dynamical equations following from an