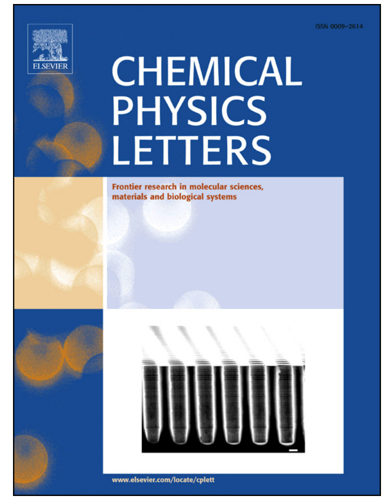

Short-Lived Electron Transfer in Donor-Bridge-Acceptor Systems

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Short-Lived Electron Transfer in Donor-Bridge-Acceptor Systems

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Abstract

We investigate time-dependent electron transfer (ET) in benchmark donor-bridge-acceptor systems. For the small bridge sizes studied, we obtain results far different from the perturbation theory which underlies scattering-based approaches, notably a lack of destructive interference in the ET for certain arrangements of bridge molecules. We also calculate wavepacket transmission in the non-steady-state regime, finding a featureless spectrum, while for the current we find two types of transmission: sequential and direct, where in the latter, the current transmission *increases* as a function of the energy of the transferred electron, a regime inaccessible by conventional scattering theory.

Keywords: tunnelling, electron transfer, quantum wires, non-perturbative methods

1. Introduction

Electron transfer (ET) in donor-bridge-acceptor (D-B-A) systems composed of organic molecules or molecular chains has been studied actively for the past few decades due to the potential applications to molecular electronics, DNA, etc. Expressions for estimating ET rates, closely-related to those derived for the superexchange mechanism [1], specifically applied to ET by the McConnell expression [2], have found extensive application especially for biological systems [3, 4, 5] and they continue to be widely used for such systems (*e.g.* photosynthesis, proteins, DNA) [6, 7, 8, 9, 10, 11].

The Landauer formula states that the electronic conductance through a molecule connected to two leads is proportional to the transmission, a quantity which depends on the scattering processes involved. Some of the methods used to describe the scattering include Green's function methods, transfer matrix approaches [12], or the use of Lippmann-Schwinger scattering operators for mapping the non-equilibrium system onto an equilibrium one [13]. All of these methods are time-independent, thus enabling the use of the Landauer or, in the case of several leads, the Landauer-Büttiker formula, for evaluating the current.

Most studies of electron transmission through bridges, *e.g.* Ref. [14], are in fact based on non-equilibrium Green's function methods rather than explicitly time-dependent calculations. Recently however, time-dependent approaches are increasingly being applied. For instance the wave-packet dynamics

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