Electronic Relaxation and Diffusion on Dynamically Disordered Lattices and Nanoparticles: Decoherence and Dissipation

by

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Declaration

I hereby declare that the work reported in this Thesis is entirely original. This Thesis is composed independently by me at Raman Research Institute under the supervision of Prof. Reji Philip. I further declare that the subject matter presented in this Thesis has not previously formed the basis for the award of any degree, diploma, membership, associateship, fellowship or any other similar title of any university or institution.

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Certificate

This is to certify that the Thesis entitled **Electronic Relaxation and Diffusion on Dynamically Disordered Lattices and Nanoparticles: Decoherence and Dissipation** submitted by Navinder Singh for the award of the degree of Doctor of Philosophy of Jawaharlal Nehru University is his original work. This has not been published or submitted to any other University for any other Degree or Diploma.

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SYNOPSIS

This Thesis is divided in two parts.

Part I: is concerned with theoretical studies on the effects of dissipative coupling to the environmental degrees of freedom on certain quantum phenomena that strictly have no classical analogue. These include (a) Quantum diffusion on a lattice described by a tight-binding one band Hamiltonian involving tunneling between the sites (orbitals) coupled dissipatively to the the environmental degress of freedom, (b) Orbital diamagnetic motion of a charged particle moving on a three-center-annulene linking Aharonov-Bohm(A-B) flux and coupled to the environment dissipatively, (c) The system (b) on a continuous ring. While in (a) and (b) the dissipative coupling is treated phenomenologically using certain Lindblad operators that project on to the lattice sites, the treatment (c) is based on dissipative coupling treated with the Feynman Path Integrals.

Part II: is concerned with the theoretical investigation of the classical kinetics of the non-equilibrium distribution of non-degenerate gas of electrons photoexcited far from equilibrium in the metallic and the semiconducting samples in the presence of dissipative coupling to the bath---the phonons .

The questions posed and the answers obtained in the Thesis are summarized below.

PART I :

(a): The Lindblad operators are known to cause unphysical heating up of the system towards infinite temperature, even as they maintain the physical characteristics of the reduced density matrix , namely, its complete positivity, hermiticity, and the trace-class nature. The question, therefore, is whether or not we can still obtain physically meaningful diffusive quantum motion of a particle with the dissipative coupling to the environment realized through the lindblads . Our answer comes out in the affirmative under the condition that the system be band-width limited as in the case of a one-band tight-binding lattice Hamiltonian with the band-width much less than $k_B T$. The latter is readily realized in the Wannier-Stark superlattices . We have obtained the mean-squired displacement as also the mean displacement, with and without the biasing field respectively. Also , with a time-harmonic drive , we have obtained a resonant enchancement of the diffusion coefficient.

PART I: (b) and (c) The question was if the orbital diamagnetic motion of a charged particle on a (b) discrete , or (c) a continuous ring linking an Aharonov- Bohm (A-B) flux gets suppressed (decohered) so as to kill orbital diamagnetism which is known to be a purely quantum effect . The question is all the more significant as the earlier treatments for the orbital motion in a plane perpendicular to the magnetic field gave the orbital moment as the decreasing function of the dissipative coupling , an effect over and above the temperature effect. In these systems the charge particle directly experiences the Lorentz force. In contrast to this , in our model systems the magnetic field enters only through the quantum phase --- geometric /topological. Indeed , our answer for the ring systems in question is that the orbital moment is not suppressed by the dissipative coupling , except for the temperature effects(heating) . Our treatment for the case (b) is based on the Lindblad coupling (phenomenological), while for the case (c) it is based on the Feynman path integral using the Euclidean action and the Caldeira-Leggett model of the dissipative coupling to the bath of harmonic oscillators.

PART II:

The stochastic model for the dissipative granular gas has been generalized to the case of the relaxation of the distribution for a non-equilibrium non-degenerate gas of photo- excited electrons in a semiconducting sample . Analytical results have been obtained by us for the steady -state under continuous (cw) optical pumping, and the boundary condition of infinitely fast recombination across the band-gap. The distribution shows a single peak structure. Our generalized treatment holds for the full range of parameters involved. Thus it goes beyond the Two-Temperature model well known in the literature on photoexcited systems. We have also treated the relaxation of the distribution function for the case of multi-phonon processes where the phonon-bath temperature enter through the time -scale that involves the electron-phonon coupling and the phonon temperature. Finally , we have given an explicit calculation of the electron-surface-phonon interaction that dominates the relaxation process for nanometric scale particles.

Publications arising out of the thesis:

In refereed journals

(1) Dissipative electron-phonon system photo-excited far from equilibrium
Navinder Singh and N. Kumar, JSTAT L06001(2005).

(2) Quantum diffusion on a dynamically disordered and harmonically driven lattice with static bias: Decoherence
Navinder Singh and N. Kumar, Mod. Phys. Lett. B, Vol. 19, 379(2005).

(3) Relaxation of femtosecond photoexcited electrons in a polar indirect band-gap semiconductor nanoparticle
Navinder Singh, Pramana ---journal of physics, Vol. 64, 111(2005).

(4) **Relaxation between electrons and surface phonons of a homogeneously photoexcited metal film**
Navinder Singh, Pramana---Journal of physics, Vol. 63, 1083(2004).

(5) Hot electron relaxation in a metal nanoparticle: Electron surface-phonon interaction
Navinder Singh, Mod. Phys. Lett. B, Vol. 18, 1261(2004).

(6) Relaxation of femtosecond photoexcited electrons in a metallic sample
Navinder Singh, Mod. Phys. Lett. B, Vol. 18, 979(2004).

In Symposia:

(1) Quantum diffusion on a dynamically disordered and driven lattice with static bias
Navinder Singh , N. Kumar, DAE Solid State Physics Symposium, GNDU/ Amritsar, Dec 26- 30, 2004 (poster).

(2) Relaxation between hot electrons and surface-phonons in small metal particles
Navinder Singh, STATPHY 22, July 4-9, 2004/ I.I.Sc , Bangalore (poster).

(3) Relaxation of femtosecond photoexcited electrons in a metallic sample
Navinder Singh, DAE-BRNS National Laser Symposium. I.I.T / Kharagpur, Dec 22-24, 2003 (poster)

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