LAGRANGIAN HOMOGENISATION OF QUANTUM-WELL SUPERLATTICES FOR NONLINEAR OPTICS

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ABSTRACT

A unitary mathematical framework that incorporates all scales in a single theory is based on a Lagrangian description of the interaction between electromagnetic fields and matter. By judicious choice of trial functions, allied with multiple-scales asymptotic ordering, an effective medium theory is constructed in which the parameters of Lagrangian trial functions evolve according to effective Maxwell equations, with medium parameters that can be identified as effective nonlinear susceptibilities whose explicit forms can be read off directly from the Lagrangian fomulation. In this theory the microscopic dynamics of the medium is described by Maxwell-Bloch equations in an appropriate Lagrangian form. The spatial averaging necessary for homogenisation is supplied automatically by the integration of Lagrangian densities which is intrinsic to Lagrangian formulations.

INTRODUCTION

Semiconductor quantum-well superlattices form a class of metamaterials whose properties for nonlinear optics are very desirable. They can be grown by standard epitaxial processes such as molecular beam epitaxy (MBE), and their optical properties can be engineered on a spatially-selective basis by means of quantum-well intermixing.

Although the microscopic dynamics of these materials is well understood, being completely described by quantum-mechanics, it is a nontrivial problem to translate the microscopic models into an effective macroscopic medium theory, because of the presence of nonlinearity, and spatial and temporal dispersion associated with resonances in the components of the medium. A series of spatial scales has to be successively homogenised, from the fundamental scale of bond dipoles (a few angstroms), the electron and hole confinement scales (a few nanometres), the superlattice periodicity (∼10 nm), to the optical wavelength (of the order 1 μm).

A unitary mathematical framework that incorporates all scales in a single theory is based on a Lagrangian description of the interaction between electromagnetic fields and matter. By judicious choice of trial functions, allied with multiple-scales asymptotic ordering, an effective medium theory is constructed in which the parameters of Lagrangian trial functions evolve according to effective Maxwell equations. The generality of this Lagrangian description, here applied to quantum-confined media, suggests that Lagrangian homogenisation can be made into a completely general procedure applicable to the homogenisation of any complex medium with arbitrary patterns of microscopic inhomogeneity, including nonlinearity, provided that suitable trial functions can be constructed.

LAGRANGIAN FORMULATION

The Lagrangian for the interaction of an electromagnetic field with a medium described quantum-mechanically is as follows. Define an *action functional*

$$S = \int_{-\infty}^{\infty} L \mathrm{d}t \tag{1}$$

and a *Lagrangian*

$$L = \int_{\mathcal{R}^3} \Lambda \mathrm{d}^3\mathbf{x} \tag{2}$$

where Λ is the *Lagrangian density* having physical units of energy density (J m^{-3}). The Lagrangian can be

expressed in terms of the various fields by the following expressions:

$$L = L_{\rm em} + L_{\rm qm} + L_{\rm int} \tag{3}$$

where

$$\begin{aligned} \Lambda_{\rm em} &= \tfrac{1}{2}\epsilon_0(\mathbf{E}\cdot\mathbf{E} - c^2\mathbf{B}\cdot\mathbf{B}) && (4)\\ \Lambda_{\rm qm} &= \tfrac{1}{2}\mathrm{i}\hbar(\psi^*\partial_t\psi - \psi\partial_t\psi^*) - \psi^* H_0\psi && (5)\\ \Lambda_{\rm int} &= \mathbf{E}\cdot\mathbf{P} = \mathbf{A}\cdot\mathbf{J} - \phi\rho. && (6) \end{aligned}$$

The current density $\mathbf{J}$ and charge density ρ are defined from the quantum mechanical wavefunction ψ by

$$\mathbf{J} = (e\hbar/m)\,\mathrm{Im}\{\psi^*\boldsymbol{\nabla}\psi\}, \quad \rho = -e|\psi|^2.$$

The electric and magnetic fields may be expressed in terms of potentials

$$\mathbf{E} = -\boldsymbol{\nabla}\phi - \partial_t\mathbf{A}, \qquad \mathbf{B} = \boldsymbol{\nabla}\times\mathbf{A} \tag{7}$$

with a gauge condition

$$\boldsymbol{\nabla}\cdot\mathbf{A} + c^2\partial_t\phi = 0. \tag{8}$$

The Lagrangian is subjected to variations in the potentials $\mathbf{A} \to \mathbf{A} + \delta\mathbf{A}$, $\phi \to \phi + \delta\phi$ and the quantum wavefunction $\psi \to \psi + \delta\psi$, resulting in the Euler-Lagrange equations $\delta S = 0$:

$$\boldsymbol{\nabla}\times\mathbf{H} = \partial_t\mathbf{D} + \mathbf{J}, \qquad \boldsymbol{\nabla}\cdot\mathbf{D} = \rho \tag{9}$$

for the electromagnetic fields, and

$$\mathrm{i}\hbar\partial_t\psi = H\psi \tag{10}$$

for the wavefunction ψ of the electronic states in the material, where H is the Hamiltonian operator of the quantum-mechanical system in position representation.

The Lagrangian can also be expressed in the transform domain by

$$S = (2\pi)^{-1}\int_{-\infty}^{\infty}\tilde{L}\,\mathrm{d}\omega, \quad \tilde{L} = (2\pi)^{-3}\int_{\mathcal{R}^3}\tilde{\Lambda}\,\mathrm{d}^3\boldsymbol{\kappa}, \quad \tilde{L} = \tilde{L}_{\rm em} + \tilde{L}_{\rm qm} + \tilde{L}_{\rm int}. \tag{11}$$

The Lagrangian (3) is a complete microscopic specification of the system, and the Euler-Lagrange equations are Maxwell's equations coupled to the Schrödinger equation (10).

MACROSCOPIC TRIAL FUNCTIONS

For definiteness, we consider the problem of plane wave incidence from a bulk semiconductor medium onto a superlattice of quantum wells of well width b and period d, under conditions of significant nonlinear optical response in the superlattice due to electronic resonances in the h1-e1 complex of subband levels. The objective of homogenisation in this instance is to describe the interaction between the macroscopic potential $\mathbf{A}$ and polarisation-current layers confined to each quantum well, in such a way that the superlattice layer appears to be replaced by an equivalent layer of homogeneous material with macroscopic properties. Suppose first that the incident wave is polarised parallel to the plane of the wells. Then there is no dipole polarisation perpendicular to the wells, only a current density $\mathbf{J}_\parallel$ flowing in the plane of the well due to the motion of the electrons. This current density is itself determined by the potential driving the electron motion, and may be due either to bound or free electrons, depending on whether the electron has enough energy to escape the valence bonding in the solid. The current density $\mathbf{J}$ exists only in a very thin layer of width b. When acting as a source for the fields in the MQW region, this can be treated as effectively a sheet current of infinitesimal width $\mathbf{J} = \mathbf{K}\delta(z)$ with $\mathbf{K} = b\mathbf{J}$ when the sheet is located at $z = 0$. Coupling these currents in layers $z = ld, l \in \mathcal{Z}$, as sources for the wave equation, leads to the dispersion law for EM Floquet waves in a periodic medium of repeated quantum wells each separated by a distance d.

The quantum expression for the determination of electron momentum $\mathbf{p}$, and hence current $\mathbf{J}$, is

$$\mathrm{i}\hbar\partial_t\langle\mathbf{\Xi}\rangle = \langle[\mathbf{\Xi}, H]\rangle \tag{12}$$

where $\mathbf{\Xi}$ is the operator for momentum, $\mathbf{p} = \langle\mathbf{\Xi}\rangle$, $\mathbf{J} = -Ne(\mathbf{p} + e\mathbf{A})/m$, and H is the Hamiltonian (energy) operator. The Hamiltonian in the presence of applied field is given by

$$H_{ss'}(\mathbf{k}_\parallel) = E_s(\mathbf{k}_\parallel)\delta_{ss'} + \frac{e}{m}\mathbf{A}\cdot\mathbf{\Xi}_{ss'}(\mathbf{k}_\parallel), \tag{13}$$

where s and s' are the indices for quantum-well eigenstates. The matrix elements $\mathbf{\Xi}_{ss'}$ belong to the momentum operator for the discrete states of the quanum well. We simplify this to the case of only 2 discrete quantum-well eigenstates, a valence subband $s = 1$ and a conduction subband $s = 2$. Near the zone centre, the energy levels are approximated by $E_s(\mathbf{k}_\parallel) \sim E_{s0} + \hbar^2\|\mathbf{k}_\parallel\|^2/2m_s$, where m_s is the effective mass of the subband with index s, and the dependence of the momentum coupling matrix elements on $\mathbf{k}_\parallel$ can be neglected. If only one component of $\mathbf{A}$ couples to the quantum-well electrons, then the Hamiltonian is approximated near the zone-centre by

$$H_{ss'}(\mathbf{k}_\parallel) = \{E_{s0} + \hbar^2\|\mathbf{k}_\parallel\|^2/2m_s\}\delta_{ss'} + \mathrm{i}(-1)^s\frac{e}{m}A_y p_0\delta_{s\,3-s'}, \tag{14}$$

where p_0 is a momentum scale constant. The density matrix for this system satisfies

$$\mathrm{i}\hbar\partial_t\Gamma(\mathbf{k}_\parallel) = [H(\mathbf{k}_\parallel), \Gamma(\mathbf{k}_\parallel)] \tag{15}$$

and expectations are computed from

$$\langle X\rangle = \int_{\mathrm{BZ}_\parallel}\mathrm{Tr}\{\Gamma(\mathbf{k}_\parallel)X\}\mathrm{d}^2\mathbf{k}_\parallel. \tag{16}$$

The Liouville equation (15) is a standard 2-level problem for each $\mathbf{k}_\parallel \in \mathrm{BZ}_\parallel$. 2-level atomic evolution by the Maxwell-Bloch equations is well-known to include optical nonlinearity due to the saturation in energy absorption by a 2-level transition [1]. The linear properties of the medium described in this way for ω far from resonance are represented by the dispersion law for EM Floquet waves

$$\frac{4}{d^2}\sin^2\frac{\beta d}{2} \quad = \quad \kappa_0^2\left\{n^2 + \frac{e^2p_0^2}{4\pi^2\epsilon_0\hbar m^2\omega^2 d}\int_{\mathcal{BZ}_\parallel}\left[\frac{1}{\Omega_{21}(\mathbf{k}_\parallel) - \omega} + \frac{1}{\Omega_{21}(\mathbf{k}_\parallel) + \omega}\right]\mathrm{d}^2\mathbf{k}_\parallel\right\} \tag{17}$$

with $\kappa_0 = \omega/c_0$ under conditions $n\kappa_0 d << \pi$, where n is the refractive index of the background host material. This is an example of homogenisation of the dielectric properties of the quantum-well layer for the case $n\kappa_0 d << \pi$, assuming a simple microscopic model for the electron motion. If $\beta d << \pi$ also, then the left-hand side of (17) is approximately equal to β^2; the dispersion law is then identical to that which would be obtained by imagining the current in the quantum wells to be smoothly distributed throughout the interwell regions. However, this situation requires that the second term in brackets on the right-hand side be small enough; unfortunately, this term contains resonances and therefore may be large.

It is known that a time-harmonic sheet current

$$\mathbf{J}(t) = \tfrac{1}{2}\mathbf{u}_y K_l\delta(z - ld)\exp(-\mathrm{i}\omega t) + \mathrm{c.c.}$$

radiates plane waves in both directions

$$\mathbf{A}_l(z,t) = \tfrac{1}{2}\mathbf{u}_y A_l\exp(\mathrm{i}\kappa_z|z - ld| - \mathrm{i}\omega t) + \mathrm{c.c.} \tag{18}$$

with $A_l = -\frac{1}{2}\mathrm{i}\mu_0\kappa_z^{-1}K_l$. The complex amplitudes of the sheet currents are generally nonlinear functionals of the total potential present at the site of the sheet $z = ld$. Also, as a consequence of the nonlinear response of the quantum-well to an applied field, harmonic generation of each time-harmonic component of the incident field occurs. It is therefore necessary to represent the radiated potentials as superpositions of all harmonics of the fundamental frequency ω :

$$\mathbf{A}_l(z,t) = \tfrac{1}{2}\mathbf{u}_y\sum_{r=-\infty}^{\infty}A_{lr}\exp(\mathrm{i}\kappa_r|z - ld| - \mathrm{i}r\omega t) \tag{19}$$

where the additional index r refers to the harmonic frequency. For self-phase modulation (SPM) and self-induced transparency (SIT) interactions, all harmonics can be neglected if they are not phase-matched, and the only significant interactions occur within the fundamental ($r = \pm 1$).

The coefficients A_{lr} are variational parameters, and the Lagrangian is a function of all these coefficients. The current in each quantum well at $z = ld$ can be described by

$$\mathbf{K}_l(t) = \tfrac{1}{2}\mathbf{u}_y \sum_r K_{lr} \exp(-\mathrm{i}r\omega t), \tag{20}$$

$$K_{lr} = F_{lr}(A_{11}, \ldots, A_{l'r'}, \ldots). \tag{21}$$

If this functional relation can be established explicitly from the microscopic dynamics, then the two Lagrangian terms $\Lambda_{\mathrm{int}} + \Lambda_{\mathrm{qm}}$ can be combined to a single term

$$\Lambda_{\mathrm{int}} + \Lambda_{\mathrm{qm}} = \Lambda_{\mathrm{pol}} = \int_0^{\mathbf{A}} \mathbf{J}(\mathbf{A}) \cdot \mathrm{d}\mathbf{A} \tag{22}$$

whose variational derivative with respect to the potential $\mathbf{A}$ is just equal to $\mathbf{J}$. These substitutions make the action functional S a function of all the variational parameters A_{mr}, with the part arising from Λ_{em} in particular being quadratic in these variables. The variational parameters may now now be interpreted as spatial samples of a fictitious spatially bandlimited macroscopic potential $\bar{\mathbf{A}}(\mathbf{x})$, and the variational problem rewritten in terms of this macroscopic potential. The Euler-Lagrange equations of this macroscopic variational problem are macroscopic Maxwell equations for the homogenised superlattice material. This procedure is a generalisation of the average Lagrangian method of Whitham [2]. In the linear case the functionals F_l are linear, and the system can be solved straightforwardly; if the wells are identical and equally spaced, this yields exactly the dispersion law (17) for Floquet plane waves $A_l = A_0 \exp(\mathrm{i}\beta ld)$.

As an example, the nonlinear dispersion relation derived by this method for a macroscopic plane wave propagating through a multiple quantum-well layer under conditions of nonresonant SPM ($\omega << \Omega_{21}(\mathbf{0})$) is

$$\frac{4}{d^2} \sin^2 \frac{\beta d}{2} = \kappa_0^2 \left\{ n^2 + \chi^{(1)}_{\mathrm{QW}}(\omega) + \chi^{(3)}_{\mathrm{QW}}(\omega) |E_0|^2 + \ldots \right\} \tag{23}$$

with $E_0 = \mathrm{i}\omega A_0$, and with explicit expressions for the order-n susceptibilities $\chi^{(n)}_{\mathrm{QW}}(\omega)$.

CONCLUSION

A Lagrangian method for homogenisation of multiple nonlinear layers into a uniform macroscopic medium has been proposed, and illustrated on the example of multiple parallel quantum-wells with a simple quantum-electronic model for the electron dynamics in the quantum wells. The method is completely general, however; the result of the method is a set of effective Maxwell equations for the dynamics of a macroscopic field which is actually fictitious, but is the best bandlimited approximation, in the Lagrangian sense, to the microscopically varying field in the interior of the medium. The example considered in the text reduces the quantum dynamics to an ensemble of 2-level systems parameterised by the in-plane Bloch wavevector $\mathbf{k}_{\|}$. This is adequate for odd-order nonlinearities such as SPM and SIT. For even-order nonlinearity, such as second-harmonic generation, three energy levels must be involved at each $\mathbf{k}_{\|}$ [3].

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