Non–linear supersymmetric σ–Model for Diffusive Scattering of Classical Waves with Resonance Enhancement

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We derive a non–linear σ–model for the transport of light (classical waves) through a disordered medium. We compare this extension of the model with the well–established non–linear σ–model for the transport of electrons (Schrödinger waves) and display similarities of and differences between both cases. Motivated by experimental work (M. van Albada et al., Phys. Rev. Lett. 66 (1991) 3132), we then generalize the non–linear σ–model further to include resonance scattering. We find that the form of the effective action is unchanged but that a parameter of the effective action, the mean level density, is modified in a manner which correctly accounts for the data.

The supersymmetric non–linear σ–model (SNSM) is a powerful tool for the description of electron transport through disordered media. In compact form, SNSM contains information on diffusion, reflection and transmission, including non–perturbative aspects like localization and parametric correlations. The purpose of this letter is twofold. (i) We extend SNSM to the transport of light (classical waves) through disordered media. The extension shows that SNSM is not restricted to Schrödinger waves but applies equally to classical waves of sufficiently high frequency. We use the Ward identities to check our results. These identities differ in form significantly for classical and for Schrödinger waves. Although the effective action of the SNSM is the same in both cases, the Ward identities are fulfilled because the source terms differ. (ii) We generalize SNSM further by allowing for both diffusive and resonance scattering. We do so with the help of random matrix theory. The generalization is motivated by an experiment on resonant light scattering. The generalization leaves the form of the effective action unchanged but endows a parameter of the action (the mean level density) with an energy dependence which correctly accounts for the data. As a result, we obtain a unified description of diffusive and of resonant scattering for both Schrödinger waves and classical waves.

We first derive SNSM for classical waves and then turn to the generalization involving resonance scattering. Following common practice, we replace vector fields by scalar quantities and consider the scalar wave equation

\[ |\Delta + k^2 \epsilon(r)| \Phi = 0 \tag{1} \]

where \( k \) is the wave number. The dielectric constant \( \epsilon(r) \) is the sum of the background dielectric constant \( \epsilon_0 \) (a constant) and the fluctuating part \( \delta \epsilon(r) \) which represents disorder. We assume that \( \delta \epsilon(r) \) is a Gaussian random process with vanishing first moment and a second moment given by

\[ < \delta \epsilon(r_1) \delta \epsilon(r_2) > = \frac{4\pi}{\ell k d+1} \delta(r_1 - r_2), \tag{2} \]

where \( \ell \) is the elastic mean free path (connected to the diffusion constant \( D \) and the energy transport velocity \( v_E \) via \( D = (1/d) v_E \ell \)), and \( d \) is the dimension of the system. It is well to recall the difference between the scalar wave equation and the Schrödinger equation for an electron with energy \( E \) in a disorder potential \( V \). Formally, the quantity \( k^2 \epsilon_0 \) in Eq. \( (1) \) corresponds to \( E \), while \( k^2 \delta \epsilon \) plays the role of \( V \). We note, however, that \( k^2 \delta \epsilon \) is energy dependent while \( E \) is not. This is the fundamental difference between classical and Schrödinger waves.

Following Efetov, we calculate the supersymmetric generating functional for the average of the product of a retarded and an advanced Green function for the differential operator in Eq. \( (1) \). We mention in passing that John and Stephen derived a non–linear sigma model for classical waves. This derivation was confined, however, to waves of fixed energy and thus bypassed the crucial issue of correlations between amplitudes at different energies. Let \( k_1, k_2 \) be the \( k \)–values of the two Green functions. With \( k_0^2 = (k_1^2 + k_2^2)/2 \) and \( \Delta k^2 = k_1^2 - k_2^2 \), the effective action is given by

\[ \mathcal{L}[Q] = \int dr \left( \frac{\pi\nu}{8\tau} \text{trg} Q^2 + \frac{1}{2} \text{trg} \log \left[ k_0^2 + \frac{\Delta k^2}{2} L + \Delta + i\eta L - \frac{1}{2\pi \tau} Q \left( 1 + \frac{\Delta k^2}{2k_0^2 L} \right) \right] \right). \tag{3} \]

In order to facilitate a comparison with Efetov’s expression for electrons, we have introduced the density of states \( \nu \) per unit of \( k_0^2 \) and per unit of volume, and the quantity \( \tau = k_0^{-2} \ell/(2\pi\nu) \) which is formally equivalent to the mean free time. We use the notation of ref. \( (3) \). The symbol \( \text{trg} \) denotes the supertrace. The supermatrices have dimension eight. The matrix \( L \) is given by \( \text{diag}(1, 1, -1, -1, 1, 1, -1, -1) \). The term \( \Delta \mathcal{L} = (1/2\tau)QL(\Delta k^2/2k_0^2) \) is due to the
energy dependence of the scattering potential $k^2 \delta \epsilon$ and would be absent in the case of electrons. Except for this term the effective action formally agrees with the one derived for electrons. The non–linear $\sigma$–model is generically derived with the help of a saddle–point condition that is valid for weak disorder, $k_0 \ell \gg 1$. Comparing the two terms proportional to $\Delta k^2$ in Eq. (3), we see that for weak disorder, the term $\Delta \mathcal{L}$ can be neglected. Then, $\mathcal{L}[Q]$ is identical to the case of electrons. The saddle–point condition has the solution $Q = iL$. Expanding around the saddle point and integrating over the massive modes, we obtain exactly the same form for the effective action as in the case of electrons. Hence, there is no difference between the non–linear sigma models for Schrödinger waves and for classical waves. This is the first important result of our work. It obviously extends to the generating functionals of all higher correlation functions and, thus, applies universally. The actual differences between the two theories are due to the different forms of the source terms.

The Ward identities for classical and for Schrödinger waves differ substantially \cite{1}. Because of the frequency dependence of the $k^2 \delta \epsilon$ term in Eq. (1), there occurs an additional term for classical waves which is absent for Schrödinger waves. How can identical effective actions give rise to different Ward identities? This is due to the difference of the source terms of both theories. The Ward identities are indeed fulfilled in both cases \cite{1}.

We turn to the generalization of SNSM including resonance scattering. To motivate both the problem and our modeling, we recall the experiment by van Albada et al. \cite{2}. For light scattering, disorder is often produced artificially: A powder is immersed into some liquid. Random scattering occurs if the dielectric constants of powder and liquid are sufficiently different. In the experiment of ref. \cite{2}, the powder used (TiO$_2$) consisted of grains with a size distribution centered around a diameter of 220 nm. For such grains, a Mie resonance occurs close to the wavelength $\lambda \sim 630$ nm of the laser light used in the experiment, causing a resonance enhancement of the scattering. This enhancement led to unusually low values of the diffusion constant $D$. Indeed, with $D$ determined from the intensity autocorrelation function versus frequency of the transmitted light, and the elastic mean free path $l$ determined from weak localization (enhanced backscattering), or from the dependence of the transmitted intensity on the length $L$ of the disordered slab, the relation $D = 1/3 \, v_E l$ yielded a value $v_E = (5 \pm 1) \times 10^7$ m s$^{-1}$ for the energy transport velocity $v_E$ through the disordered medium. This value is about an order of magnitude smaller than the phase velocity.

This surprising result has been understood both for small concentration \cite{7} and, more recently, also for strong concentration \cite{12} of the scatterers. (In the latter case, the resonant structure in the diffusion constant versus frequency disappears, and an overall decrease of $D$ is observed \cite{13}). Qualitatively speaking, the transport velocity is reduced because on its way through the medium, the energy is stored for some time in the Mie resonances.

Our generalization of the SNSM goes beyond this work in two ways. First, it yields a unified theoretical framework in which both average and fluctuation properties can be calculated on the same footing. This is in contrast to previous approaches \cite{14,15} which use the Bethe–Salpeter equation \cite{2} or a mean–field approximation \cite{13} for the calculation of the transport velocity $v_E$, and a diagrammatic impurity perturbation expansion for the intensity autocorrelation function. Second, the generalized SNSM identifies the energy dependence of the mean level density $\rho(E)$ as the culprit for the observed deviation from standard behavior. A simple argument which yields the same result as the analytical derivation and illuminates the physical content may be helpful at this point. In the case of electrons \cite{16}, the Thouless formula $g = E_c/\Delta$ connects the average conductance $g$ with the Thouless energy $E_c = hD/L^2$ and the mean level density $\rho(E) = 1/\Delta$. Here, $L$ is the length of the sample. The presence of numerous resonances with equal resonance energies $E_1$ leads to a local Breit–Wigner–shaped increase of $\rho(E)$ near $E_1$. Since $g$ is not affected by the presence of the resonances, the Thouless formula implies that $E_c$ and, hence, the diffusion constant have a Breit–Wigner–like dip near $E_1$. Because of the equality of the effective action in the SNSM for Schrödinger and for classical waves, this argument applies likewise to scattering of light. With increasing concentration of scatterers, the dip widens and becomes less deep. Eventually, this results in an overall decrease of $D$ over a wide frequency interval. We note that in the context of SNSM, the relation $D = 1/3v_E l$ is not used explicitly. It is replaced by the Thouless relation.

To account for the presence of resonances, we proceed as follows. For the quasi one–dimensional geometry appropriate in the present context, we use the well–known identity between SNSM and a band random matrix model \cite{17}. In the framework of the latter, it is easy to model the presence of additional resonances. With standard supersymmetry techniques, we finally map the resulting generalized band random matrix model back onto SNSM. This yields the required generalization. The procedure is the same for classical and for Schrödinger waves.

In the absence of resonances, a quasi one–dimensional disordered system of length $L$ for diffusive scattering of electrons or light is modeled \cite{17} as consisting of many longitudinal slices of length $l_0 \ll L$. Within each slice, the Hamiltonian is modeled as a matrix $H_{GOE}$ of dimension $N$ belonging to the GOE, the random–matrix ensemble with orthogonal symmetry. Neighboring slices are coupled by Gaussian–distributed uncorrelated random matrix elements. The strength of this coupling defines the diffusion constant $D_0$. The first and the last slice are coupled to the channels, i.e., the asymptotic states for free propagation of electrons or light. The transmission through the disordered region
is given in terms of the squares of elements of the scattering matrix which connect incident and outgoing channels on either side of the disordered region.

To account for the presence of Mie scatterers, we modify the form of the Hamiltonian $H$ within each slice. Now, $H$ is a matrix of dimension $N + m$ where $m$ is the number of Mie scatterers within that slice. The value of $m$ is given by the concentration of scatterers, and by the linear dimensions of the slice. In an $(N, m)$ block representation, $H$ has the form

$$H = \left( \begin{array}{cc} H_{GOE} & V \\ V^T & E_1 \times I_m + H_{res} \end{array} \right).$$

Here $T$ denotes the transpose. The matrix $H_{GOE}$ of dimension $N$ was introduced in the previous paragraph, $I_m$ is the $m$-dimensional unit matrix, $E_1 = \hbar \omega_1$ is the resonance energy of each of the $m$ Mie scatterers with equal resonance frequencies $\omega_1$, the rectangular matrix $V$ couples the $m$ resonances to $H_{GOE}$, and the random matrix $H_{res}$ describes the coupling between the $m$ resonances. The matrices $V$, $H_{res}$, and $H_{GOE}$ are uncorrelated. The second moment $(H_{GOE})_{\mu \nu} (H_{GOE})_{\mu' \nu'}$ of $H_{GOE}$ is given by $(\lambda^2/N)(\delta_{\mu, \mu'} \delta_{\nu, \nu'} + \delta_{\mu, \nu'} \delta_{\nu, \nu'})$. Here, $\lambda$ determines the average level spacing $\Delta_S$ in each slice. For $m = 0$, we obtain Wigner’s semicircle law for the mean level density, typical for random matrix theory. We assume the energy $E_1$ to lie at the center of the semicircle, $E_1 = 0$ where $\Delta_S = \pi \lambda / N$. The matrix $H_{res}$ is also a member of a GOE with a second moment of the same form as for $H_{GOE}$ but with $\lambda$ replaced by $\lambda_1$. The strength factor $\lambda_1$ is chosen in such a way that the interaction between resonances results in a lifting of the degeneracy which is of the order of the mean level spacing $\Delta_S(0)$ so that $\lambda_1 \ll \lambda$. Too strong an interaction would wash out the resonance structure altogether. Without loss of generality, the rectangular matrix $V$ can be taken to have non-zero elements $v$ only on the main diagonal $\nu = m$. Since all scatterers are assumed to be identical, all these elements are taken to be equal. It is possible to estimate $\lambda_1$ and $v$ microscopically from the properties of the Mie resonance, and from the concentration $\mu$ of scatterers. The calculation is not done here. Suffice it to say that $\lambda_1$ grows more strongly than linearly with $\mu$.

Before we cite complete results, it is instructive to consider the case of a single slice. After averaging and the Hubbard–Stratonovich transformations relating to the two different statistical ensembles $H_{GOE}$ and $H_{res}$, two supermatrices are introduced, leading to two coupled saddle–point equations. These are solved easily. The resulting mean level density has the form

$$\rho(E) = \frac{N}{\lambda \pi} + m \frac{(\Gamma + b \lambda_1)/\pi}{(E - (E/2\lambda) - a + \lambda_1^2 + (\Gamma + b \lambda_1)^2},$$

Here $\Gamma = v^2/\lambda$ is the width of the Lorentzian in the absence of interresonance coupling, and $a + ib = \sigma_1$ is the solution of the second saddle-point equation $\sigma_1 = \lambda_1/(E - \lambda_1 \sigma_1 - \Gamma E/2\lambda - i \Gamma)$. The last term shows the resonance enhancement of $\rho$, centered at the energy $E_1 = 0$ of the $m$ resonances. The total width $(\Gamma + b \lambda_1)$ is determined by both, the strength $\lambda_1$ of the interresonance coupling and the strength $v$ of the coupling to the random scatterers. For small $\lambda_1$ (corresponding to a low value of the concentration $\mu$), we regain the expression for uncoupled $(\lambda_1 = 0)$ resonances given in ref. 13, whereas in the limit of high concentrations $(\lambda_1 \gg \Gamma)$ the Lorentzian peak in $\rho(E)$ disappears (see Fig. 1). It is easy to check that $\rho(E)$ obeys the requirement $\int \rho(E) \, dE = N + m$. The calculation of the two–point function suffers from the presence of two small parameters $E - E_1$ and $\lambda_1$ in addition to $\Delta k^2$. However, under the realistic condition $\Gamma \gg \Delta_{eff} = (\rho)^{-1}$, the remaining supersymmetric integral attains a simple form $\Delta_{eff}$: The effective action coincides with the case without resonance scattering (pure GOE) except for a rescaling of the average level spacing, i.e. the replacement $\pi \lambda / N \to \Delta_{eff}$. All resonance effects follow from this rescaling. Additional differences to the pure GOE case can occur only in the wings of the resonance in Eq. (1), or in cases where $\Gamma \sim \Delta_{eff}$.

Using standard supersymmetry, we show that this result carries over to the case of a quasi one–dimensional sample consisting of many slices. The effective action of the resulting SNSM has Efetov’s form,

$$\mathcal{L}_{eff} = -\frac{\pi}{8V \Delta_{eff}} \int \text{tr} \left[ D_{eff}(\nabla Q)^2 + 2i \Delta k^2 L Q \right] \, dr,$$

where the effective diffusion constant $D = D_0 \Delta_{eff}/\Delta$ is given in terms of the diffusion constant $D_0$ and the mean level spacing $\Delta$ in the absence of resonances, and in terms of the effective mean level spacing $\Delta_{eff}$ due to the presence of resonant scatterers, and $V$ is the volume of the system. Eq. (8) implies that the transmission through a sufficiently long sample is unaffected by the presence of resonances while all correlation functions as well as $D$ do depend on the effective mean level spacing $\Delta_{eff}$ and, thus, show a strong dependence on the resonances.

To compare with experiment, we note: (i) For classical waves in a uniform medium in $d$ dimensions, the non–resonant part of the density of states is proportional to $\epsilon^{d/2}$. Increasing the number of resonating spheres, we increase
the effective dielectric constant of the medium. Hence, \( \lambda \) should decrease with \( \mu \). (ii) As pointed out above, \( \lambda_1 \) is expected to increase monotonically with \( \mu \). Both these trends affect the dependence on concentration of \( \Delta_{\text{eff}} \) and, therefore, of \( D \sim \Delta_{\text{eff}} \). With increasing \( \mu \), point (i) causes a decrease of \( D \) far away from the resonance, and point (ii) a widening of the resonance. Fig. 1 shows \( D \) versus energy for different concentrations. In agreement with the experimental results [3,4], we find a deep dip in the diffusion constant at the resonance energy. This dip is caused by the resonance enhancement of \( \Delta_{\text{eff}} \) and is particularly pronounced at low concentrations of the resonant scatterers. As the concentration increases, the dip is smeared out. At the same time, the value of \( D \) outside the resonance decreases. We stress that the presence of \( H_{\text{res}} \) in the Hamiltonian is crucial for the agreement with experiment.

In summary, we have shown that for sufficiently high frequency, the effective action in the supersymmetric non-linear \( \sigma \)–model for diffusive scattering of classical waves is identical in form with the analogous expression for electron transport in disordered media. This statement holds in spite of the fact that the Ward identities differ. Moreover, we have generalized the supersymmetric non-linear \( \sigma \)–model to include resonance scattering. This generalization leaves the form of Efetov’s effective action unchanged but endows the parameters with an energy dependence of Breit–Wigner form. As a result, the transmission remains unchanged but correlation functions and diffusion constant show a strong resonance dependence. This is in agreement with experiment. The results are also valid for electrons and therefore could apply to future experiments on mesoscopic samples.

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FIG. 1. Normalized diffusion constant $D/D_0$ as a function of $(E - E_1)/\Delta S(0)$ for different concentrations of the resonances ($D_0$ is the diffusion constant of the medium in the absence of resonances).