Mesoscopic Conductors and Correlations in Laser Speckle Patterns

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Submicrometer disordered metallic systems at low temperatures display interesting conductance fluctuation effects, owing to the interference of coherent electron wave transmission through the system. This quantum interference phenomenon also gives rise to the striking experimental observation that the $1/f$ noise power (where f is frequency) in disordered conductors increases as the temperature is decreased, contrary to common intuition. The theoretical techniques developed for this problem can be applied to the transmission of light through a random medium, and this yields novel predictions for the correlations in the laser speckle patterns and the possibility of studying the positions and motions of scatterers in a medium which multiply scatter the probing light.

THE STUDY OF ELECTRICAL CONDUCTIVITY AND OTHER transport phenomena is an old subject in solid-state physics. The conventional approach deals mainly with macroscopic samples (~1 mm in size) at room temperatures. In this case the sample conductivity is determined only by the average scattering rates at which lattice vibrations (phonons), other electrons, and impurity atoms scatter a forward-moving electron out of its direction of motion (that is, its Bloch state **k**). The motion of the electron is usually treated within the semiclassical Boltzmann transport theory. The justification for such a simple approach is the following: At room temperature, inelastic scattering by phonons occurs at a very high rate, for instance, of the order $1/\tau_{in} \sim k_B T/\hbar \sim 10^{13}\ s^{-1}$, and since the phase information of the electron wavefunction is destroyed each time it is scattered inelastically, the typical distance at which an electron remains phase-coherent is very short, roughly of the order ~100 Å. Thus in this high-temperature regime, any effects associated with interference of coherent electron wavefunctions are confined to this short length scale, and further, they are relatively unimportant in describing macroscopic electron transport. Crudely speaking, the electrons here can be regarded as semiclassical particles, and their wave aspects (which necessarily leads to the interference effects) can be approximately ignored.

With rapid advances in microfabrication technology in recent years, it is now possible to produce in the laboratories metal or doped-semiconductor samples with dimensions on the order of or smaller than 1 μm. When such small samples are cooled down to low temperatures (for instance, <1 K), the wave coherence length of the electron wavefunction L_Φ becomes the order of or even longer than the sample dimensions L. In this regime, it becomes crucial to include the phase coherence of the wavefunctions properly in describing the transport properties of system, and the semiclassical Boltzmann equation approach is inadequate. This regime has come to be known in the field of condensed matter physics as the mesoscopic regime, which means that owing to phase coherence of the electron wavefunction the system displays physical behaviors that are in between the familiar macroscopic semiclassical picture and one that requires an atomic or molecular description (*1*).

We shall concentrate our attention in this article on the disordered mesoscopic metal systems, that is, we assume that our conducting samples contain a significant amount of impurity atoms or structural disorder. Our discussion is also valid for doped semiconductors, for such systems at low enough temperatures behave in the same way as a metal, albeit with a much slower Fermi velocity v_F. The strength and the concentration of the impurities in the system can be characterized by the elastic scattering mean free path l, which physically represents the typical distance over which the wavevector **k** of an electron in a Bloch state becomes significantly altered. Typically, l is of the order 100 Å, and it is important to notice that it does not depend on the temperature. This is because elastic scattering does not involve any change in the electron's energy. Another crucial point is that elastic impurity scattering only has the effect of changing the simple Bloch states of an electron (with a well defined **k**) into a much more complicated multiply scattered wavefunction, but it does not affect the electron wavefunction's phase coherence. A useful, if overly simplified physical picture for electron transport in a disordered metal sample is one in which an electron undergoes random walks because of multiple scattering with the impurity atoms, with a step size l, while maintaining phase coherence up to time τ_{in}. The random walk leads to a diffusion constant $D = v_F l/3$, and an expression for the phase coherence length $L_\Phi = \sqrt{D\tau_{in}}$. Since the inelastic scattering rate $1/\tau_{in}$ generally decreases with the temperature T, for a sample of size ~1 μm, the mesoscopic regime is reached when the temperatures are below 1 K.

As we shall now see, the interference among the multiply scattered electron partial waves leads to many novel fluctuation phenomena in mesoscopic conductors. But first we shall introduce the Landauer formula, which ties together the problem of mesoscopic conductance fluctuations and the seemingly unrelated problem of the correlations effects in laser speckle patterns, both of which we shall discuss in the present article.

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The Landauer Formula for Quantum Conductance

The conductance of a disordered metal in the mesoscopic regime $L < L_\Phi$ can be related to the transmission probability of the electronic wavefunction propagating from the left of the disordered metal sample (see Fig. 1A) to the right, via the Landauer formula (*2*):

$$G = \frac{e^2}{h} \sum_{ab} T_{ab} \tag{1}$$

where T_{ab} is the intensity transmission coefficient at Fermi energy ϵ_F from a propagating waveguide mode a in the (clean) left lead to the sample to the b mode on the right. These waveguide modes in the leads are also referred to as conducting "channels," and they essentially refer to the various quantized directions at which an incoming electron wave is incident onto the disordered metal sample (and similarly for the outgoing waves). The total number of such channels is given by $N \approx (k_F W)^{d-1}$, where k_F is the Fermi wavevector, d is the dimension of space, and W is the width of the sample. Equation 1 is perhaps more familiar in the context of an electron tunneling through a thin insulating barrier sandwiched between two pieces of clean bulk metals, in which case the conductance is also proportional to the tunneling transmission coefficient for the electron wave to go through the barrier. The summation over all the propagating channels in the leads refers to the physical notion that in a conductance measurement, only the total electron flux, that is, the current, is collected; and since all channels are equally populated owing to Fermi statistics in the leads, all the conducting channels contribute to quantum conductance equally. We emphasize that the Landauer formula defined this way is valid only in the mesoscopic regime $L < L_\Phi$, since the sample region $0 < z < L$ is assumed to be described by wavefunction transmission with full phase coherence. The power of such a relation is that it reduces the problem of quantum conductance, or mesoscopic transport, to that of a scalar wave propagation through a disordered scattering medium. Thus quantum transport is just like a scattering experiment in nuclear or elementary particle physics, where the disordered conductor plays the role of a big "nucleus." It is also instructive to note that the parameter e^2/h, which has the dimension of conductance (or inverse resistance), $[e^2/h \approx (25.8\ k\Omega)^{-1}]$, sets the natural scale for all the quantum transport problems, since the wavefunction transmission coefficients are dimensionless quantities. This is the fundamental reason that e^2/h enters in an essential way in all the well known examples of quantum transport, such as electron localization (*3*), the quantized Hall effect (*4*), and the recently discovered quantized longitudinal conductances in quasi-one-dimensional systems (*5*). For example, in a metal with conductivity σ, the average conductance $\langle G \rangle$ equals $\sigma W^{d-1}/L$ by Ohm's Law, where d is the dimension of space (thus, for thin films $d = 2$). We can easily show, using the Boltzmann expression for σ, that $\langle G \rangle = (e^2/h)(Nl/L)$. The dimensionless number $g \equiv \langle G \rangle/(e^2/h)$ plays a central role in the modern theory of the electron localization problem, (*3*), and it has been shown that the system is metallic only if g exceeds unity. This is the regime that we shall consider in this article.

We also note that from this point on, the transmission coefficients are no longer specific to the problem of quantum transport in mesoscopic metals, where the transmission coefficients refer to that of a coherent propagating electron wavefunction going through a disordered conductor at very low temperatures. In fact, they can refer to any coherent scalar wave propagation through a disordered scattering medium, such as a laser light (in the scalar wave approximation) propagating through a thick slab of glass filled with small voids ("air bubbles"); or an ultrasonic wave propagating through a piece of metal which contains microcracks (again in the scalar approximation). The only replacement of parameters necessary to make our discussion applicable to the case of light or sound is simply $k_F \leftrightarrow k = \omega/c$, and the understanding that l should be taken as the transport mean free path of light (or sound) l^*. This is the reason why many of the techniques developed in the mesoscopic conductance fluctuations can be readily applied to study the various correlations and fluctuations effects in a general wave propagation problem in the multiple scattering regime, such as light propagation through a milky fluid or a thick scattering wall. Therefore in our discussions below, we shall use the language of electron transport in mesoscopic metals and that of coherent light transmission through a slab containing random scatterers interchangeably.

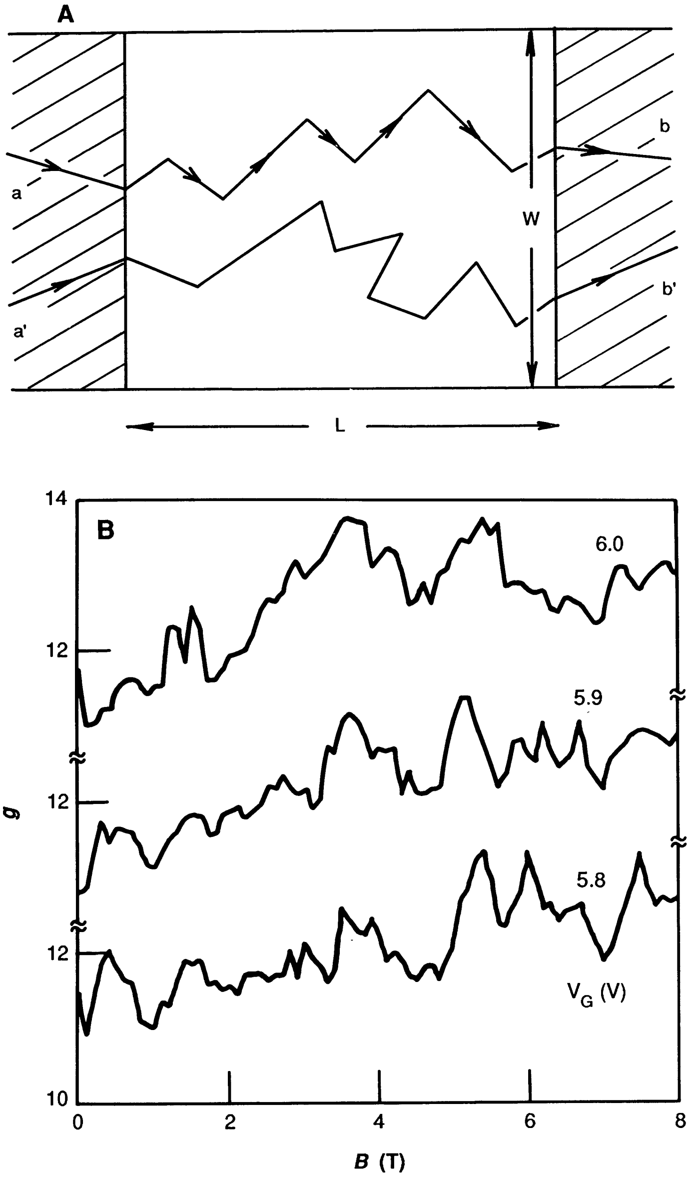


Fig. 1. (**A**) Geometry for studying conductance fluctuations in mesoscopic disordered conductors, and also for understanding the Landauer conductance formula which maps the conductance problem into that of a general wave propagation through random scattering media. The shaded regions are the ideal leads which are free of impurities (or scatterers in the optical context). A conductor is mesoscopic if its size L is less than the phase coherence length L_Φ. (**B**) A typical measurement of conductance versus magnetic field illustrating the phenomenon of universal conductance fluctuations, done on a mesoscopic doped semiconductor sample, taken from Skocpol *et al.* (*6*). The vertical axis $g = G/(e^2/h)$ is the dimensionless conductance. The three curves are effectively for three different value of the Fermi wavevector k_F. Note that the fluctuation in g is of order unity.

Universal Conductance Fluctuations

The average conductance attributable to disorder scattering is given reasonably accurately by the Boltzmann theory. The first hint of the importance of quantum interference effects comes from considering the fluctuations of the conductance. It has been recently demonstrated, both experimentally (*6*) and theoretically (7), that a mesoscopic conductor exhibits the so-called "universal conductance fluctuations." This phenomenon can be summarized as follows: For mesoscopic conductors with $L \leq L_\phi$, the conductances vary from sample to sample, (that is, for different realizations of the locations of the impurity atoms), so that its root-mean-square fluctuations $\delta G \equiv \sqrt{\langle (G - \langle G \rangle)^2 \rangle}$ is roughly given by e^2/h. Here the average $\langle\ \rangle$ refers to an average over an ensemble of similar mesoscopic conductors with different realizations of the impurity positions. As we saw in the last section, e^2/h is the fundamental conductance unit for quantum transport, so it is not too surprising that it sets the size of conductance fluctuations in a mesoscopic conductor. We note that the size of these "universal conductance fluctuations" are anomalously large, when viewed from the standpoint of semiclassical transport theory. In the latter case one can estimate the magnitude of fluctuations by considering the sample as a collection of independent grains of linear dimension l, so that one expects $\delta G_c/\langle G \rangle \sim (l/L)^{d/2}$ for a cubic sample ($L = W$). Since the average conductance is given by $\langle G \rangle = \sigma L^{d-2}$, this implies that the semiclassical expectation for sample-to-sample conductance fluctuations is $\delta G_c \sim e^2/h\ (l/L)^{-(4-d)}$, that is, it goes down with the size of the system L with a power $(4 - d)/2$. Thus we see that the universal conductance fluctuations represent enhanced conductance variations from sample to sample owing to quantum interference, when the phase coherence length L_ϕ is sufficiently large at low temperatures.

Experimentally, these sample-to-sample conductance fluctuations are actually observed in one given sample, which is however subject to a varying applied magnetic field. As the magnetic field adds a phase factor to the electron partial waves between the various random scattering events, it has the same effect as changing the positions of the impurity atoms randomly, thus effectively allowing one to change from one sample to another. The amount of magnetic field change that has to be imposed to effectively change a sample can be estimated from the following simple argument: The area covered by two typical interfering scattering paths is roughly LW (see Fig. 1A and consider for simplicity $L = W$). We know from quantum mechanics that, if a magnetic field change is such that the flux change enclosed by these two paths, $\Delta\phi \sim (\Delta B)LW$, exceeds the flux quantum $\phi_0 = hc/e$, then these two paths will acquire a phase difference of $\sim 2\pi$ and we would then have effectively created a new sample by this change of the applied magnetic field. Thus the measured magnetoconductance (see Fig. 1B) should exhibit random fluctuations with a typical field scale of $\Delta B_c \approx \phi_0/(LW)$. This is indeed seen in experiments (*6*). We also need to emphasize that these seemingly "random" magnetoconductance fluctuations for a given mesoscopic conductor as a function of the applied field can be differentiated from noise. When one changes the field up and down, the fluctuating magnetoconductance from our mechanism should be completely reproducible, provided that no impurity motion has occurred during the experiment. Thus this magnetoconductance curve can be regarded as a kind of "magneto-finger print" of the given disordered conductor. But what can we say about the conductance fluctuations when just such impurity motion occurs?

Sensitivity of the Mesoscopic Conductance to Impurity Motions: Implications for 1/*f* Noise

So far we have established that for a mesoscopic conductor $L \leq L_\phi$, the conductance changes by an amount $\sim e^2/h$ when the positions of all the impurity atoms are altered in a random way, that is, when we go from one sample to another. It is natural to ask what the conductance fluctuations would be if we only move one impurity. The physical relevance of such a question is the following: At the very low temperatures where mesoscopic effects are important, the impurity atoms are usually frozen in their positions; in other words, the probability for them to be thermally activated to hop to another site is prohibitively low. (Such a probability is $\sim e^{-\Delta E/k_B T}$, where the typical energy barrier for impurity hops in a metal is of the order ~ 0.1 eV.) But impurity atoms can still hop due to quantum tunneling events. Since a mesoscopic sample is very small, estimates suggest that even such quantum impurity hops are not very frequent. So it is relevant to consider theoretically what the typical conductance change is when just one impurity atom makes a hop, which we denote as δG_1, observable in mesoscopic conductors when a single impurity atom undergoes a quantum tunneling event to move to another metastable position. Without a calculation, one might make the naïve guess that just like in semiclassical theory, one gets $\delta G_1 \approx \sqrt{1/N_i}\,\delta G \approx \sqrt{1/N_i}\,(e^2/h)$, where $N_i = n_i L^d$ is the total number of impurities in the system, and n_i is the impurity concentration. A detailed calculation indicates instead that (*8*)

$$\delta G_1^2 \approx \frac{1}{N_i}\frac{L^2}{l^2}\,\delta G^2 \qquad (2)$$

Thus we see that the effect of moving a single impurity on the typical conductance change is enhanced by a large factor L^2/l^2. Physically, this result can be understood from an argument involving the diffusive interference processes. Going back to Fig. 1A, we see that the typical number of scattering events by a particular electron partial wave propagating across the mesoscopic sample is on the order of $(L/l)^2$. Thus the probability that a given simple impurity is visited by this typical scattering path should be the ratio of the total scattering volume of this path $(L/l)^2\sigma_0 l$ to the volume of the entire system L^d, where σ_0 is the scattering cross section of a given impurity. The motion of this impurity will change the phase of the scattering path visiting it by a random amount, so that the ratio between δG_1^2 and δG^2 should be given by this probability, and we arrive at our result Eq. 2, after using the relation between the elastic mean free path and the scattering cross-section $1/l = n_i\sigma_0$. An especially interesting and somewhat surprising special case is a thin film sample of thickness t (that is, $d = 2$), where

$$\delta G_1^2 \approx \frac{\sigma_0}{lt}\left(\frac{e^2}{h}\right)^2 \qquad (3)$$

thus the typical conductance change induced by moving just one impurity is a constant fraction of the conductance change induced by moving all the impurities, which is independent of the size of the film, as long as $L \leq L_\phi$. This shows how sensitive the conductance of a given sample is to the motion of a single impurity.

When one considers the question of the typical conductance change caused by moving m impurity atoms, one can show theoretically that $\delta G_m^2 \approx m\delta G_1^2$, as long as $(m/N_i)(L^2/l^2) \leq 1$, and for the opposite case δG_m saturates at $\delta G \approx e^2/h$, which corresponds to the typical conductance change due to moving all the impurities. We now note the interesting result that in order to obtain the maximum quantum conductance fluctuations $\delta G \approx e^2/h$ for a mesoscopic conductor, one does not need to move all the impurity atoms. Rather, it suffices to move only a small fraction $p \equiv m/N_i$ of the impurities, when $p \approx l^2/L^2$, which is usually a small percentage. This is another illustration of the extreme sensitivity of mesoscopic conductance to impurity motions (*8*).

This sensitivity of quantum conductance to impurity motions allows the possibility of using a simple conductance measurement in small metal wires or films to study the quantum tunneling events of an individual or a group of impurity atoms at low temperatures.

Such experiments have been performed recently (*9*), and it shows rather good agreement with the theoretical predictions from the picture just outlined. More recently, this novel feature of mesoscopic conductance fluctuations has been used to study experimentally the spin dynamics in a small size metallic spin-glass system, where the very long time scale spin fluctuations can be probed rather sensitively by measuring the magneto-conductance fluctuations of the system at low temperature (*9a*).

This theory of mesoscopic conductance fluctuations caused by impurity motions can be used to predict the magnitude of the $1/f$ noise in disordered metals at relatively low temperatures (say <70 K) where the multiple scattering condition $l \ll L_\phi$ is satisfied. (This corresponds to a low temperature regime where the average conductivity of the sample is no longer temperature dependent.) Furthermore, the theory is applicable even to macroscopic samples when $L \gg L_\phi$. This is done by assuming that a fraction p of impurity atoms can hop around randomly during the experimental measuring time, and that a large sample (a thin film for instance) can be thought of as made up of a collection of phase coherent sheets of size L_ϕ. Each such sheet must be treated quantum mechanically, so that its conductance fluctuations as a function of time are given from the theoretical considerations above, by $\delta G_\phi^2 \approx p(L_\phi^2/l^2)(e^2/h)^2$ when $p < l^2/L_\phi^2$ or $\delta G_\phi^2 \approx (e^2/h)^2$ when $p > l^2/L_\phi^2$. The electron transport between the various L_ϕ size sheets can be combined as if they were classical resistors; (that is, no phase coherence between them needs to be included); for the integrated $1/f$ noise magnitudes this gives

$$\delta G^2 = \delta G_\phi^2 \left(\frac{L_\phi}{L}\right)^2 \tag{4}$$

This result is qualitatively different from its semiclassical counterpart, and even quite contrary to common intuition: as the temperature is lowered, L_ϕ in general increases; so that the noise magnitudes, actually increase with a decreasing temperature! This is contrary, for example, to how the (white) thermal Nyquist noise behaves as a function of temperature: $\delta V^2 \approx k_B T$. Nonetheless, just such unusual temperature dependences of the noise power at relatively low temperatures has been found experimentally by a number of groups after the proposal of our theory, leaving little doubt about the essential correctness of the quantum interference interpretation for the low temperature $1/f$ noise in disordered metals (*10*).

Universal Conductance Fluctuations and Laser Speckle Patterns

How can we understand the origin of the rather counter-intuitive result of the universal conductance fluctuations from the wave propagation point of view? This question has led us to investigate the correlations among wave transmission coefficients through a disordered scattering medium. From Eq. 1, it follows that the problem of computing sample to sample conductance fluctuations reduces to the following quantity:

$$\langle(\delta G)^2\rangle = \left(\frac{e^2}{h}\right)^2 \sum_{aba'b'} C_{aba'b'} \tag{5}$$

where $C_{aba'b'} = \langle \delta T_{ab} \delta T_{a'b'} \rangle$ is the correlation function among individual transmission channels, and $\langle\ \rangle$ denotes an average over an ensemble of samples with different impurity configurations. Thus the statement of universal conductance fluctuations amounts to showing $\Sigma_{aba'b'} C_{aba'b'} \approx 1$ for any values of $l \ll L < L_\phi$ and in any dimension d. We note that in a light scattering experiment, a laser wave is incident in a given direction (channel) and the transmitted light intensity can be measured in any direction (in far field), so that the individual T_{ab} can be measured directly for a given sample. The correlation function $C_{aba'b'}$ can then be constructed experimentally by collecting T_{ab} for different samples. Thus we see that light scattering experiments offer much more detailed information on the multiply scattered wave correlations and fluctuations, and they can be used as a way to test theoretical concepts which were originally constructed for low-temperature electron transport. As we will see, this connection between these two sets of problems has also led to important new understanding about light propagation in multiple scattering random media, as well as some interesting potential applications involving the use of the speckle-pattern correlations to retrieve useful information about the properties of the scattering medium itself. Here the term speckle pattern refers to the complicated interference pattern in the transmitted intensities T_{ab} as a function of the outgoing direction b (or the light frequency) which are readily observable by the eye in any complex scattering problems. We clarify here that l stands for the transport mean free path of the scattering of light, which measures the typical distance at which the incident beam is scattered significantly out of the forward moving direction.

The intensity-intensity correlation function $C_{aba'b'}$ can be computed theoretically in the multiple scattering regime $l \ll L$, by means of a perturbative approach in which the disorder is treated as a small parameter, that is, we assume that $1/k_F l \ll 1$. The intensity transmission coefficient T_{ab} for a given sample is given by $T_{ab} = |t_{ab}|^2$, where t_{ab} refers to the complex amplitude transmission coefficients. From the path integral picture of quantum transmission, the t_{ab}'s can be represented as a sum over the amplitudes corresponding to all the possible multiple scattering paths p propagating through the sample, that is,

$$t_{ab} \approx \sum_p A_{ab}^p \tag{6}$$

where $A_{ab}^p \approx \exp(2\pi i s_{ab}^p/\lambda)$, and s_{ab}^p is the total path length of the random walk path p, with the constraint that the incoming wave is in channel a and the outgoing one is in channel b (See Fig. 1A). As $C_{aba'b'}$ involves the correlations among four such sums over all possible paths, the understanding of this function looks like quite a formidable task at first. But because most of the correlations among the different paths reduce to zero upon ensemble averaging $\langle\ \rangle$, we can obtain the most important contributions to this correlation function by identifying the subset of paths which will yield nonvanishing correlations. In Fig. 2A we draw the most obvious subset of correlated diffusion paths, which turns out to give rise to the largest contribution to the correlation function $C_{aba'b'}$, and can be evaluated to yield (*11*)

$$C_{aba'b}^{(1)} = \langle T_{ab}\rangle \langle T_{a'b'}\rangle \delta_{\Delta q_a, \Delta q_b} F_1(\Delta q_a L) \tag{7}$$

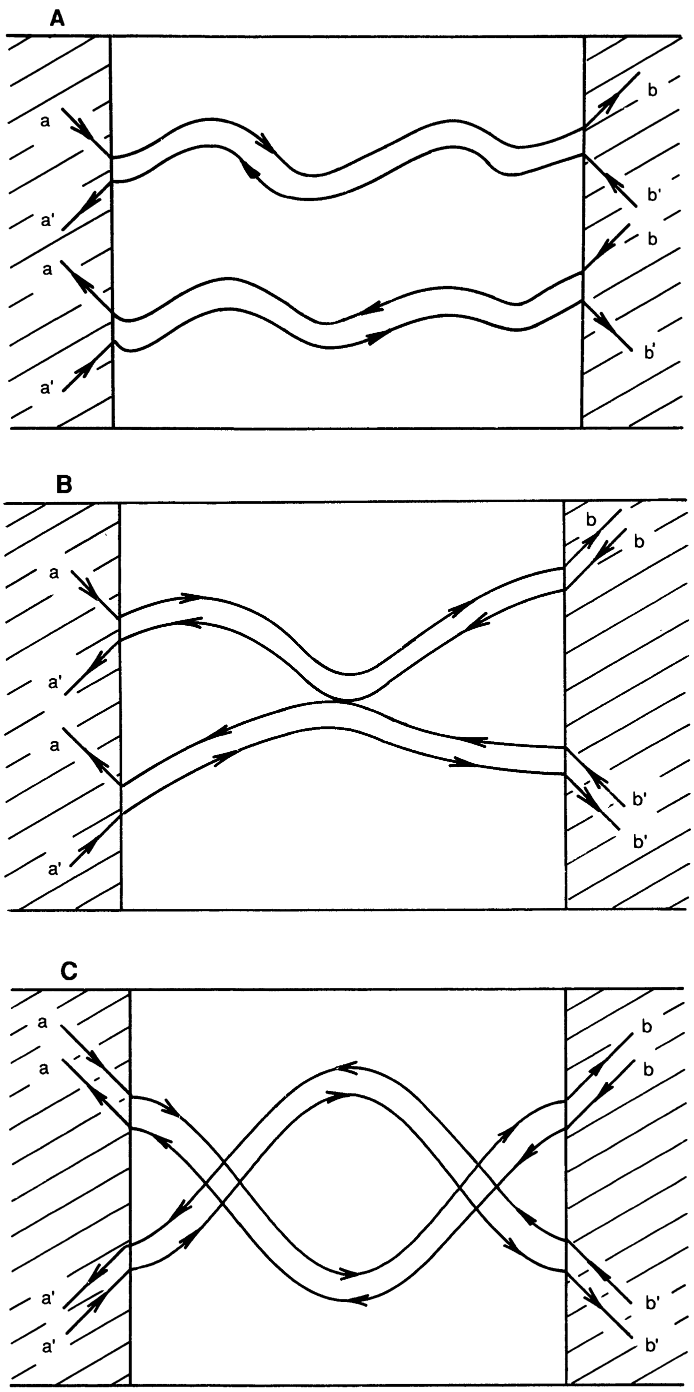
where $\Delta \mathbf{q}_a = \mathbf{q}_a - \mathbf{q}_{a'}$ and $\mathbf{q}_a$ is the component of the wavevector of the incoming wavefunction perpendicular to the z-direction, $F_1(x) = x^2/\sinh^2 x$, and $\langle T_{ab}\rangle \approx l/NL$ is the ensemble average value of the transmission coefficient.

When we set $a = a'$ and $b = b'$, Eq. 7 reduces to the relation

$$\langle(\delta T_{ab})^2\rangle = \langle T_{ab}\rangle^2 \tag{8}$$

which is a well known result in laser speckle patterns, and shows that the relative intensity fluctuations are of order unity (*12*). In fact, this is the physical reason why multiply scattered light interference patterns look so "speckle"-like.

The δ-function factor in Eq. 7 represents what we term the "memory effect." Since this $C^{(1)}$ term vanishes identically if $\mathbf{q}_a - \mathbf{q}_{a'} \neq \mathbf{q}_b - \mathbf{q}_{b'}$, and since $C^{(1)}$ is the largest contribution to the intensity-intensity correlation function, this means that a shift in the direction of the incoming beam, represented by a small $\Delta \mathbf{q}_a$, results on average in a shift of the same angle in the outgoing intensity speckle pattern (in direction space). This is quite remarkable because we are working in the multiple scattering regime $l \ll L$ so that the amount of unscattered wave going through a slab of thickness L is exponentially small, and the incoming plane wave has come through as a seemingly random speckle pattern. But if the incoming direction is changed by a small angle $\delta\theta$ less than $\delta\theta_c \approx 1/kL$ (so that the factor F_1 is not too small), it is possible to "detect" such a change by looking at the cross-correlations between the two speckle patterns. We note the important difference between this correlation angle for memory effect with the simple angular size of the individual speckles on a speckle pattern: the angular width of the speckles is governed by $1/kW$ where W is the size of the incoming beam spot (the simple diffraction limit which separates out the different "channels"). Thus by making the incoming beam size much larger than the thickness of the scattering sample, it is possible to separate the two different angular scales.



The "memory effect" has recently been verified experimentally by Freund *et al.* (*13*), whose principal results we illustrate in Fig. 3. This result can also explain the well known optical phenomenon that a speckle pattern executes apparent motions in a well defined direction as some optical component is moved (here the direction of the incoming beam). This is because the eye only notices the short term coherence (that exists in our case within the small correlation angle of $\delta\theta_c \approx 1/kL$) and ignores the longer term randomization of the pattern.

This new understanding about the correlation properties of laser speckle patterns in the multiple scattering limit may lead to potential applications in detecting moving objects behind a scattering medium whose thickness L is large compared to the transport scattering mean free path l, that is, when light or ultrasound or any other classical wave propagates through a diffusive multiple scattering medium. As indicated by the $C^{(1)}$ correlation function Eq. 7, the motion of the source at some velocity v leads directly to an average motion of the far-field speckle pattern behind the thick scattering medium by the same speed. In optical context, this allows for the possibility of performing "speckle pattern interferometry" even in the highly multiple scattering regime $L \gg l$.

It can easily be shown that upon summing over all the incoming and all the out-going channels of the $C^{(1)}_{aba'b'}$ correlation function, one obtains a corresponding variance of the conductance fluctuations which are much small than e^2/h. Mathematically, this is because $C^{(1)}$ is nonvanishing only for a very small subset of channels. Thus, to understand what correlations among the transmission coefficients are responsible for the universal conductance fluctuations, we must try to find higher order terms in the perturbation expansion of the intensity correlations which are less restricted in the range of channels where they are important. Figure 2B represents the subset of interfering paths which are responsible for the next leading order correlation process. It can be evaluated to give

$$C^{(2)}_{aba'b'} \approx \frac{\langle T_{ab}\rangle \langle T_{a'b'}\rangle}{g} [F_2(\Delta q_a L) + F_2(\Delta q_b L)] \tag{9}$$

where $F_2(x) = 2x^{-1}(\coth x - x/\sinh^2 x)$, and $g = \langle G\rangle/(e^2/h) = Nl/L$ is the Thouless parameter or the dimensionless average conductance, which in the weak disorder regime $1/kl \ll 1$ is always much larger than unity. Thus $C^{(2)}$ is much smaller than $C^{(1)}$, but it is important to note that it decays to zero only when both Δq_a and Δq_b are large. So it is nonzero for a much larger range of channels. This difference in the channel (or angle) dependence comes about because when the two sets of diffusing paths in Fig. 2B cross somewhere in the middle of the sample, two paths can exchange partners in propagating through the sample. Thus the dependence on the outgoing angular difference drops out of the correlation function. Since this can only

Fig. 2. Correlation processes for the intensity transmission coefficient correlation function $C_{aba'b'} = \langle \delta T_{ab} \delta T_{a'b'}\rangle$. Since $T_{ab} = |t_{ab}|^2$, we represent it graphically by t_{ab} and its time reversed path t^*_{ab} (with arrow reversed). Inside the sample we sum over all possible multiple scattering paths due to random scattering. The pairing of multiple scattering paths results in correlated amplitudes which do not average to zero upon ensemble average. (**A**) The correlated paths which contribute to $C^{(1)}_{aba'b'}$. (**B**) The correlated paths which contribute to $C^{(2)}_{aba'b'}$. Notice that this process is independent of the angular distance between the outgoing channels b and b'. This is the reason why $C^{(2)}_{aba'b'}$ has a nonzero limit for the case when $a = a'$ but with b being very different from b'. The crossing of the two diffusion paths gives rise to the reduction factor $1/g$. (**C**) Same as in (B), but for the correlation process $C^{(3)}_{aba'b'}$. Notice that this correlation process is independent of all the channel indices. This term, which represents effectively "infinite range" correlations among all transmission channels, is responsible for the universal conductance fluctuations in mesoscopic conductors.

happen for two sets of diffusion paths which meet once inside the system, this correlation process $C^{(2)}$ is down from the previous $C^{(1)}$ process by the probability of two diffusion paths going through the same point in space, which we have seen previously is given roughly by $1/g$. This is reminiscent of the $1/g$ factor in weak localization theory, where the quantum correction to conductance is due to paths which must self-intersect once in the system (*3*). For the case of a single incident beam, that is $a = a'$, our theory predicts that the transmitted speckle pattern intensities are correlated at large angle separations by an amount $\sim \langle T\rangle^2/g$, which is a small but constant value (*14*). This represents physically the important result that if a given speckle spot in the transmission far field is brighter than average, all the other spots also tend to be a little brighter. Thus the speckle pattern is not really very random! This is an important manifestation of the non-self-averaging nature of coherent wave transport in the multiple scattering regime. We will see that owing to its special dependence on channel separation angles, the $C^{(2)}$ correlations dominate over any correlation experiments which have a "one-channel" in, "all-channels out" type configuration.

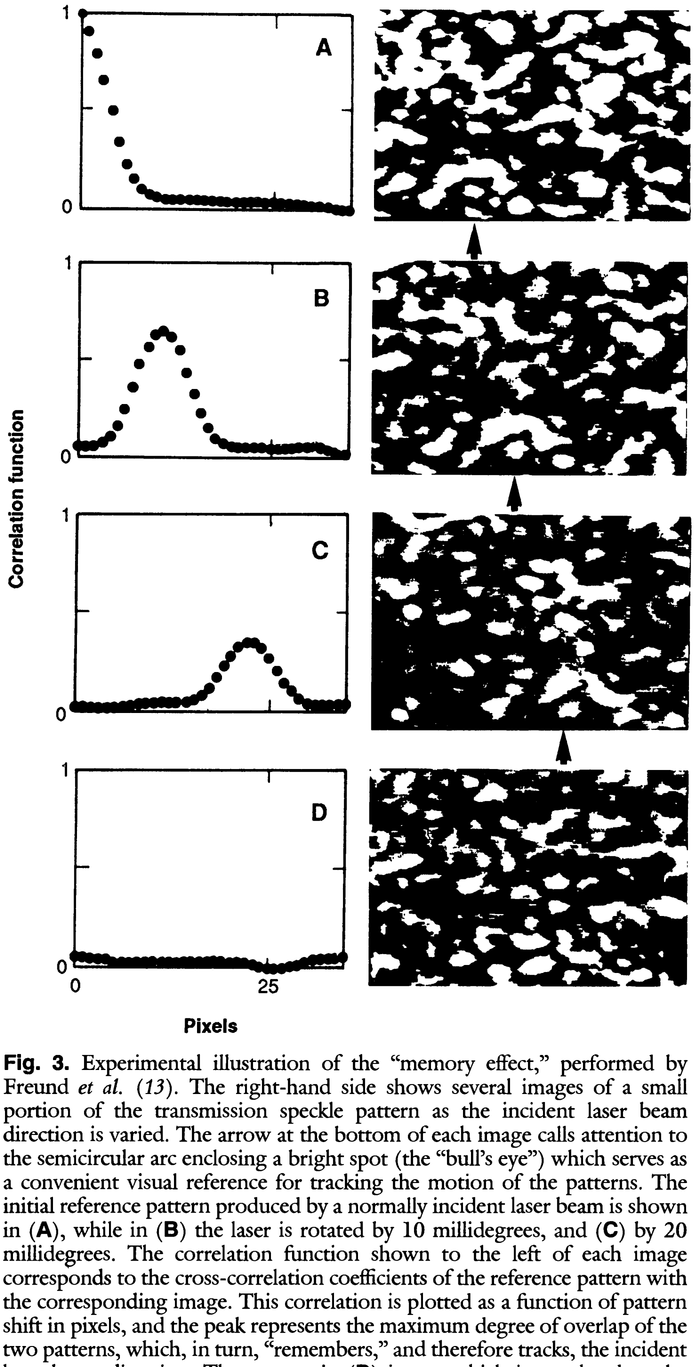


Fig. 3. Experimental illustration of the "memory effect," performed by Freund *et al.* (*13*). The right-hand side shows several images of a small portion of the transmission speckle pattern as the incident laser beam direction is varied. The arrow at the bottom of each image calls attention to the semicircular arc enclosing a bright spot (the "bull's eye") which serves as a convenient visual reference for tracking the motion of the patterns. The initial reference pattern produced by a normally incident laser beam is shown in (**A**), while in (**B**) the laser is rotated by 10 millidegrees, and (**C**) by 20 millidegrees. The correlation function shown to the left of each image corresponds to the cross-correlation coefficients of the reference pattern with the corresponding image. This correlation is plotted as a function of pattern shift in pixels, and the peak represents the maximum degree of overlap of the two patterns, which, in turn, "remembers," and therefore tracks, the incident laser beam direction. The pattern in (**D**) is one which is unrelated to the reference in (**A**), and the correlation function shows the expected small statistical fluctuations about zero. In this experiment, the various relevant parameters taken as follows: $L = 810$ μm, $W = 15$ mm, $\lambda = 0.633$ μm, and $l = 100$ μm. The absorption length is much larger than the sample thickness.

However, summing Eq. 9 over all the channel indices again yields a value for the conductance fluctuations much less than e^2/h. Thus we must go on searching for the higher order intensity correlations which are still longer ranged in the channel space. Figure 2C represents the next order correlation process, corresponding to two intersections for the two sets of paired interfering scattering paths, which can be evaluated to give

$$C^{(3)}_{aba'b'} \approx \frac{\langle T_{ab}\rangle \langle T_{a'b'}\rangle}{g^2} \approx \left(\frac{1}{N}\right)^4 \qquad (10)$$

Notice that this intensity correlation process is extremely weak. However, it has effectively "infinite" range in the channel space, or in other words all channels are correlated to each other by this small but nonzero amount. Summing this correlation function over the channel indices yields the result of the universal conductance fluctuations. Drawing analogy to the theory of weak localization, the $1/g^2$ factor reflects the fact that the fraction of random-walking paths which must intersect twice is of the order $1/g^2$.

We remark that when one considers correlations among the transmitted intensities, it does not make sense to speak generally about what correlation processes are the most dominant. This depends on which specific correlation experiment one is interested in. For example, we have seen that for individual channel intensity correlations, $C^{(1)}$ makes the largest contribution; but if one wants to consider the conductance fluctuations in mesoscopic conductors, (which amounts to a sum over all channels of the correlations among different transmission coefficients), $C^{(3)}$ is the most important correlation process. If one instead considers the correlations and fluctuations properties of the total transmitted intensity $T_a^t \equiv \Sigma_b T_{ab}$, which can also be directly measured in an optical experiment, it is easy to show that it is dominated by the $C^{(2)}$ correlation function. In other words, we can say that there are in fact three distinct kinds of speckles: the short-range, the long-range, and the infinite-range correlations; they play the dominating role in three different classes of correlation experiment: the single-channel to single-channel transmission case, the single-channel to all-channel transmission case, and the all-channel to all-channel transmission case.

It is also easy to generalize the above calculations to the case of correlations of transmitted intensities at different frequencies, for which we write the correlation function $C(\Delta\omega) \equiv \langle \delta T_{ab}(\omega)\delta T_{ab}(\omega + \Delta\omega)\rangle$. Realizing that angular shifts and frequency differences are both sources for giving rise to decorrelations of the speckle intensities, we can write down the general structure of the frequency correlations as

$$C(\Delta\omega) = C^{(1)}(\Delta\omega) + C^{(2)}(\Delta\omega) + C^{(3)}(\Delta\omega) + \ldots \qquad (11)$$

with $C^{(1)}(\Delta\omega) = \langle T_{ab}\rangle^2 \tilde{F}_1(\sqrt{\Delta\omega/\omega_c})$, $C^{(2)}(\Delta\omega) = \langle T_{ab}\rangle^2/g\ \tilde{F}_2(\sqrt{\Delta\omega/\omega_c})$, and $C^{(3)}(\Delta\omega) = \langle T_{ab}\rangle^2/g^2\ \tilde{F}_3(\sqrt{\Delta\omega/\omega_c})$, where $\omega_c = D/L^2$ is a correlation frequency $\tilde{F}_1(x)$ and $\tilde{F}_2(x)$ are approximately the same functions as the corresponding ones for describing the angular correlations, in Eqs. 7 and 10; and $\tilde{F}_3(x) \approx 1$ for $x \ll 1$, $\tilde{F}_3(x) \approx x^{-1/2}$ for $x \gg 1$. Recent experiments have measured the $C^{(2)}(\Delta\omega)$ long-range correlations, which are in excellent agreement with our theory (*15*). We remark that the frequency correlations in light experiments are the direct analogs of the Fermi energy correlations in conductance fluctuations experiments with mesoscopic conductors.

It is also quite interesting to explore the connection of the above picture with the recent development of the so called "diffusive wave spectroscopy" (DWS) (*16*). This is an area of active current research whose goal it is to exploit modern understanding about multiple scattering and apply it to the light scattering experiments in colloidal solutions, in the regime of high concentration, such that $L \gg l$, so that traditional scattering theory based on the single scattering (Born) approximation is no longer valid. From the point of view of the physics of colloidal systems, it is important to have a scattering tool that can function for high concentration systems. In this context, the stereotypical sample is a solution in which the solvent particles scatter the light, but unlike in the mesoscopic conductor problem, they are all free to move around as a result of Brownian motion. This makes the intensity transmission coefficients time dependent, that is, we have $T_{ab}(t)$.

In a DWS experiment, one usually measures the auto-correlation function

$$C(t) = \langle \delta T_{ab}(t) \delta T_{ab}(0) \rangle \tag{12}$$

It turns out for the case of the scattering particles executing simple Brownian random walks with a diffusion constant D_p (to be distinguished from the light diffusion constant D), $C(t)$ is given by a set of correlation functions having again the same structure as the angular or frequency correlations that we have considered for static scattering media, that is,

$$C(t) = C^{(1)}(t) + C^{(2)}(t) + C^{(3)}(t) + \ldots \tag{13}$$

where the largest term $C^{(1)}(t)$ takes the form

$$C^{(1)}(t) = \langle T_{ab} \rangle^2 F_1(\sqrt{t/t_0}) \tag{14}$$

where $t_0 \approx (l^2/L^2)\, \tau_0$, with $\tau_0 = 1/(D'_p k^2)$, and $F_1(x)$ is exactly the same function as in the static case, in Eq. 7, which we recall decays exponentially for large x. We see that just like in the case of mesoscopic conductances, the light intensity auto-correlations are very sensitive to the motions of the scatterers; this is made evident by the characteristic time scale t_0 for the decay of the correlation which is very much reduced from τ_0, the diffusion time of a particle over a distance given by the wavelength of the light. This effect is just another aspect of the extreme sensitivity of the coherent multiply scattered wave transmission to any motion of the scatterers in the multiple scattering regime. It allows for the possibility of probing the motions of the diffusing scatterers at very high concentrations such that interparticle interactions are important, as well as at very short time scales such that deviations of particle motions from standard Langevin random walk descriptions are possible (*17*).

The time scale t_0 for speckle decorrelation in Eq. 12 can be understood physically by considering again a typical multiple scattering path going across the sample of size L, with the number of scattering events $n \approx (L/l)^2$. After a time interval t, the scattering particles would all typically have moved a distance $\Delta r^2 \approx D_p t$. Thus the total scattering path length s_p would have been changed randomly by an amount $\Delta s_p^2 \approx nD_p t$. When t is of the order t_0, that is, when the speckle pattern has been effectively decorrelated, this should correspond to the condition $k\Delta s_p \approx 1$; that is, a phase shift of 2π for the typical scattering path, due to scatterer motions. This then leads to the expression for the time constant for the decay of the speckle pattern correlations t_0.

Similarly, we have for the higher order correlation processes $C^{(2)}(t) = \langle T_{ab} \rangle^2/g\ F_2(\sqrt{t/t_0})$, and $C^{(3)}(t) = \langle T_{ab} \rangle^2/g^2\ F_3(\sqrt{t/t_0})$. Again, for the correlations properties of the total transmitted intensities $T_a^t(t) = \Sigma_b T_{ab}(t)$, the $C^{(2)}(t)$ correlation process dominates, which we recall has a novel long-time tail in the form of $1/\sqrt{t}$. This interesting prediction on the long time tails in DWS with total transmitted intensities awaits experimental verification.

We emphasize in closing that all the above novel correlation effects among the various speckle pattern intensities in the multiple scattering regime were not contained in the conventional statistical theory of speckle patterns, which assumes the various speckle intensities to arise from a sum of statistically independent random amplitudes (*12*). Thus we see that the new insights obtained from the study of mesoscopic conductance fluctuations have served to further our understanding in a seemingly unrelated set of problems of correlations effects in multiply scattered classical wave propagations.

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