Observation of Reflectance Fluctuations in Metals

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Through the study of the power spectra of a monochromatic light beam reflected by metallic mirrors, fluctuations in their reflectance is observed. The power spectra were obtained down to a factor $10^{-6}$ below the Standard Quantum Limit, with a dynamic range of $10^5$ in the frequency and power, using methods we developed. The properties of the spectra are investigated and their dependence on the material is analyzed. The physics underlying the phenomenon is also discussed. These fluctuations provide a new window into the degrees of freedom responsible for the reflection process in metals.

I. INTRODUCTION

Reflections from surfaces, such as mirrors, are ubiquitous, and are an integral part of everyday life. In physics, studying optical properties of materials is perhaps the most powerful tool for investigating their electronic and vibrational properties [1–4]. As such, in metals, the subject has been studied for some time, and continue to be studied actively to this day [5–7]. Properties of reflection are known to depend on the wavelength of light, temperature and the material [1–4, 8], and can further depend on geometric aspects of the material, such as its size and thickness. However, fluctuations inherent in mirrors, on which we report here, seem not to have been studied so far. The problem we address may be phrased from a different intriguing perspective — can an ideal mirror yield a “perfect” reflection? Reducing this question to its simplest concrete form, if we shine a monochromatic light on an ideal metallic mirror, can we tell whether the light has been reflected or not, just from the properties of the reflected light itself? If so, can we tell by what material? The answers we find are positive for both questions. The underlying reason is that the reflection is caused by microscopic degrees of freedom, such as electrons and ion cores [3, 4]. All these degrees of freedom fluctuate both thermally and quantum mechanically, so that they affect the light, at some level. This effect should in principle be detectable, though the question remains whether this is possible within practical limits. While the fluctuations are indeed small, we have measured the fluctuations in the reflectance in metallic mirrors and found their properties to depend on the material. This opens another window into the degrees of freedom responsible for reflection in metals.

The paper is organized as follows: In Sec. II we explain the design and the realization of the experiment to measure the fluctuations in the reflectance of metals. The results obtained in the measurements are explained, and analyzed in Sec. III. The meaning of the results and their underlying physics are discussed in Sec. IV.

II. THE CONCEPT OF THE EXPERIMENT AND SETUP

When a monochromatic light beam with a constant power is shone on a flat metallic mirror, can the effects of the reflection be found in the reflected light itself? Away from the direction determined by the law of reflection, inelastic scattering effects, such as Brillouin and Raman effects, can be observed, and have proven to yield important information regarding the elastic properties of matter, as well as the electronic and vibrational properties of atoms and molecules that constitute the material [3, 4, 9, 10]. On the other hand, the reflected light is dominated by the elastically scattered light and its color is unchanged, so that we essentially have only the reflected power as its property. However, its power can depend on time, and should the microscopic degrees of freedom contributing to the reflection fluctuate, their effects should show up in this time dependence.

To measure these fluctuations, conceptually, a simple experiment can be set up as in Fig. 1. Light is shone on a mirror, and its reflection by the mirror is detected by a photodetector (PD). These fluctuations should be observable...
in the power spectrum of the reflected light power,

\[ S(f) = \int_{-\infty}^{\infty} d\tau e^{-i2\pi f\tau} \langle P(t)P(t+\tau) \rangle = \frac{1}{T} \left\langle \left| \tilde{P}(2\pi f) \right|^2 \right\rangle, \]

where \( P \) is the power of the reflected light, measured by the photodetector, and \( \langle \cdots \rangle \) indicates the ensemble average. \( T \) is the measurement time and tilde denotes the Fourier transform. Fluctuations in the reflectance is \( S_R(f) = S(f)/P^2 \), where \( P \) is the average power of the reflected light. In reality, measurements from such an implementation are dominated by the shot-noise, the random power fluctuations in light due to its discrete quantum nature, often referred to as the “Standard Quantum Limit”. The shot-noise level appears as \( 2eI \), where \( I \) is the photocurrent and \( e \) is the electron charge magnitude. It is impossible, even in principle, to separate the signal from this noise, with this kind of a simple setup.

The shot-noise appears in the same manner both for the source and the reflected light, so that no effects of the reflection process is observed in the light itself, with this method.

**FIG. 2**: Experimental configuration: Differential measurements and averaged correlations are combined to reduce the unwanted noise, such as the shot-noise and the laser noise, in the measured spectra. DM1,2 transmit light 1 and reflect light 2. Paths for the laser light 1,2 are in green, blue, respectively and paths common to laser light 1,2 are in cyan. While the light beams focused on the sample in the figure are well separated for illustrative purposes, the beams in the experiment overlap and reflect almost back along their original paths, but slightly shifted, due to the large numerical aperture (NA = 0.9) of the objective lens. Fourier transforms (FFT) and averagings of the data are performed on a computer to obtain the spectrum (inside the dashed box, red).

To uncover the effects of reflection, several obstacles need to be overcome: First, unwanted noise, including shot-noise, needs to be reduced to levels so that the fluctuations caused by the reflection become visible. Second, it needs to be established that the observed signal is not caused by the light causing changes to the mirror itself, such as damaging its surface. Third, the cause of the observed phenomenon needs to be distinguished from other possible sources of fluctuation, such as surface waves of the material.

The basic principle underlying the extraction of the spectra is to combine the differential measurements with the averaging of the correlated measurements. The former removes the light source noise, which is the same, since the source is the same. The latter reduces any noise that arises independently in the photodetector measurements, such as the shot-noise, statistically. More concretely, two light sources (laser 1,2), are used and each light is split into two and shone on two locations of the sample, as seen in Fig. 2. These two locations are the same for both light sources. Accordingly, four photocurrent measurements \( D^{(\alpha)}_j \) \((\alpha = 1,2, j = 1,2)\) are made, corresponding to the two focus locations on the sample, and the two light sources. Here, \( \alpha \) and \( j \) label the location and the light source, respectively.

Photodetector measurements have the following form;

\[ D^{(\alpha)}_j = S^{(\alpha)} + L_j + N^{(\alpha)}_j \]

\( S^{(\alpha)} \) denotes the signal, or the fluctuations, at location \( \alpha \), \( L_j \) denotes the noise in the light source \( j \) and \( N^{(\alpha)}_j \) is the shot-noise in the photocurrent \( D^{(\alpha)}_j \). To obtain the spectrum, multiple measurements of the set \( \{D^{(\alpha)}_j\} \) are taken, and the following averaged correlation is computed:

\[ \left\langle \left( \hat{D}^{(1)}_1 - \hat{D}^{(2)}_1 \right) \left( \hat{D}^{(1)}_2 - \hat{D}^{(2)}_2 \right) \right\rangle = \left\langle \left( \tilde{S}^{(1)} - \tilde{S}^{(2)} + \tilde{N}^{(1)}_1 - \tilde{N}^{(2)}_1 \right) \left( \tilde{S}^{(1)} - \tilde{S}^{(2)} + \tilde{N}^{(1)}_2 - \tilde{N}^{(2)}_2 \right) \right\rangle \]

Here, \( \langle \cdots \rangle \) denotes an averaged result. Since the fluctuations at the different locations and the shot-noise in the photocurrents are all independent from each other, their correlations all go to zero statistically, in the limit of infinite...
number of averagings. Therefore, the above averaged correlation reduces essentially to the desired spectrum,

$$\left< (\tilde{D}^{(1)}_1 - \tilde{D}^{(2)}_1) (\tilde{D}^{(1)}_2 - \tilde{D}^{(2)}_2) \right> \rightarrow \left< |\tilde{S}^{(1)}|^2 \right> + \left< |\tilde{S}^{(2)}|^2 \right> = 2 \left< |\tilde{S}|^2 \right>$$

(4)

Here, we used the property that the averaged fluctuation spectra at the two locations on the material under identical conditions, are the same, so that $|\tilde{S}^{(1)}|^2 = |\tilde{S}^{(2)}|^2 = |\tilde{S}|^2$. Since $D^{(\alpha)}$’s are photocurrent measurements, and the photocurrent is proportional to the power of light received by the photodetector, the spectrum Eq. (4) is essentially the power spectrum, Eq. (1), up to a constant. In this averaged correlation, the relative statistical error in the spectrum due to the unwanted noise is the inverse of the square root of the number of averagings. The averaging of the correlation here removes any noise that is not correlated in the two differential measurements, along with the shot-noise. It should be noted that the averaging by itself does not remove the shot-noise; if the differential measurement is averaged, we obtain, in the limit of infinite number of averagings,

$$\left< (|\tilde{D}^{(1)}_1 - \tilde{D}^{(2)}_1|)^2 \right> \rightarrow \left< |\tilde{S}^{(1)}|^2 \right> + \left< |\tilde{S}^{(2)}|^2 \right> + \left< |\tilde{N}^{(1)}|^2 \right> + \left< |\tilde{N}^{(2)}|^2 \right> = 2 \left< |\tilde{S}|^2 \right> + \left< |\tilde{N}|^2 \right>$$

(5)

Similarly to Eq. (4), the averaged noise spectrum is independent of the location, and $|\tilde{N}^{(1)}|^2 = |\tilde{N}^{(2)}|^2 = |\tilde{N}|^2$. This result contains the shot-noise that dominates the measurement, when the signal is small, which applies to the current experimental conditions. Therefore, the shot-noise level is determined more precisely with more averagings, in the measurement, Eq. (5). A conceptually simpler way to reduce the relative contribution of the shot-noise, is to increase the average light power, $\bar{P}$, since the power spectrum, Eq. (4), behaves as $\sim \bar{P}^2$ and the shot-noise behaves as $\mathcal{P}$. This, in practice, is not an effective method here — when light powers large enough to reduce the shot-noise to levels that uncover the spectrum are used, the sample itself is damaged. Furthermore, it precludes us from using smaller light powers to systematically study the power dependence of the spectrum, as is done in the next section. While the observed phenomena and the measurement systems were different, the above same basic principle, in essence, was used previously to achieve factors of $10^{-3}$ to $10^{-5}$ reduction in the shot-noise, in the measurements of surface thermal fluctuations of fluids[13, 16], and spontaneous noise in atomic vapor[17, 18].

Let us briefly mention the technical aspects of the setup used in this work (Fig. 2). Two laser sources with wavelengths 488nm (Sapphire 488, Coherent, USA) and 532nm (Samba, Cobolt, Sweden) were combined into a single beam with a dichroic mirror (DM1), then split into two beams by a beam splitter (BS). The beams were reflected at two locations of the mirror at nearly normal incidence (separation 77 μm). The light beams were focused at the mirrors down to the diffraction limit, using a microscope objective lens (Olympus MPLFLN100XBDP) with a high numerical aperture value (NA= 0.9). The reason for this is explained in the next section. The light coming into the polarizing beam splitter (PBS) from the source is horizontally polarized, which is then circularly polarized at the sample using a quarter-wave plate (QWP). The reflected light is vertically polarized by going through QWP, so that it is reflected by PBS towards the photodetectors (PD, Hamamatsu Photonics S5973-02). The reflected light powers of the beams were measured by photodetectors, whose differential measurements were digitized using analog-to-digital converters (ADC, Picoscope ps6404A). The digitized output was processed on a computer to obtain the spectrum.

III. EXPERIMENTAL RESULTS AND ANALYSIS

Results from a measurement using the methods in Sec. II are shown in Fig. 3 in which it is seen that the signal is measured down to $10^{-6}$ times the Standard Quantum Limit, with around $10^5$ factor in the dynamic range, both in the frequency and the spectral magnitude. In the figure, measurement without correlation is the averaged differential measurement, Eq. (5), and the fluctuation spectrum was obtained from the averaged correlation, Eq. (4). Here and below, the spectra were normalized using the shot-noise level, $2eI$, in the photocurrent power spectrum. The spectra $S(f)$ were normalized for the output signal of a single photodetector, and the reflectance fluctuation spectra $S_{R}(f)$ are independent of the normalization. The light beam powers applied were 8 μW to 2.5 mW at the mirror, per beam. Metal coated planar mirrors of unprotected gold (PF10-03-M03, Thorlabs, USA), unprotected aluminum (TFAN-15S03-10, Sigma Koki, Japan), and protected silver (PF10-03-P01, Thorlabs, USA) were used in the experiment.

The light beams in the experiment travel through, and are reflected by various materials, including beam splitters, a quarter wave plate, dichroic mirrors, a lens and air, apart from the sample mirror. Therefore, it is imperative to establish that the measured fluctuations arise from the reflections by the sample mirror at the two beam spots. The physics underlying this is that the beams are focused down to the diffraction limit only at the mirrors, so that the fluctuations from other components are averaged out over the beam. This is why an objective lens with a large numerical aperture was used to focus the beam to its diffraction limit at the mirror, and another reason why a setup as simple as Fig. II is insufficient. The cause of the fluctuations was also experimentally confirmed as follows: During
the measurements, the light beams from the two light sources were focused on the same beam spots. When the beams were focused on different points, while keeping the rest of the light paths still overlapped, the fluctuation spectrum disappeared, showing that the fluctuations originate from the reflections at the sample mirror. One should add that the dependence of the spectra on the sample material can only be explained by the the sample being the signal source, since the experiments are otherwise identical.

Several measurements were made at different locations of the mirror to confirm the reproducibility of the data in each case. The measurement times for the spectra were $3 \times 10^4$ s to $3 \times 10^5$ s, with more averagings, and hence longer times required for lower light powers.

The observed signal does not exist in the incoming source light and is the sign of reflection by the mirror. However, more work is needed to ascertain whether this is a property of the material or the light affecting the mirror. To this end, power spectra for the reflected light were measured for different incoming light beam powers. The results are
shown in Fig. 4 for a gold mirror. One clearly sees that the spectra are similar in shape, which indicates that the light is acting as a probe and is not affecting the mirror in an essential manner. Had the light affected the mirror, it is difficult to imagine that the spectral shape is unaffected, since the effects should grow with the power of the light beam. Also, if the intrinsic behavior of the mirror is being observed, the process should be linear, so that the power spectrum should be proportional to the square of the average light power, $P^2$. This can be seen in Fig. 4 (inset); by rescaling the spectra by $P^{-2}$, the spectra essentially become identical, showing the similarity of their shape and its dependence on the light power as $P^2$. The frequency dependence of the spectrum is well described over the whole measured range by $f^{-1.25}$, which can also be observed from the spectra in Fig. 4. To distinguish these fluctuations from traveling waves on the mirror surface, previously measured in light scattering at non-specular directions[9, 10], the spectra were measured using differences in the light power fluctuations at two close locations, separated by 77 $\mu$m. Surface waves with longer wavelengths will be correlated and eliminated in this differential measurement.

A natural question is what happens for other metals. In Fig. 5, the power spectra of the reflected light are shown for an aluminum mirror for various light beam powers and in Fig. 5 (inset), the spectra rescaled by $P^{-2}$ are shown. It is again seen that the spectral shapes are essentially independent of the light power and their magnitudes behave as $P^2$. The frequency dependence is again seen to be well described by $f^{-1.25}$, but there is a slight crossover behavior at around 200 Hz. The frequency dependence of the spectra could have been different for different metals, but interestingly enough, were similar for gold and aluminum, except for the crossover behavior that exists for aluminum at low frequencies. We also measured the power spectra for a silver mirror and found that their shapes are independent of the power and consistent with the $f^{-1.25}$ behavior.

To quantitatively analyze the spectra, we express the spectra as $S(f) = b(P)f^{-1.25}$, for $f \gtrsim 200$ Hz. The behavior of $b(P)$ with respect to the light power is shown in Fig. 6. It is found that $b(P)$ does depend on the light power as $b(P) = \alpha P^2$, as mentioned above, and $\alpha$ depends on the metal. To understand the underlying dynamics, the dependence on $\alpha$ on the absorptance, $A = 1 - R$ ($R$: reflectance), is shown in Fig. 6 (inset). $\alpha$ is seen to increase with the absorption rate, though more data is needed to establish its form of the dependence.

**IV. UNDERLYING PHYSICS AND DISCUSSION**

For optical wavelengths, the absorptance decreases with increasing free electron density[1–4], and since the statistical noise from independent objects decrease with their number, perhaps this suggests the source of the spectra. Free electrons play a most important role in reflection, and it might seem that they are responsible for the observed spectra. This is, however, unlikely for the following reasons: First, the time scales corresponding to the observed spectra are in the range 0.1 s to 0.1 $\mu$s. Free electrons in these metals have mean free times in the order of $10^{-14}$ s and transit times
in the light beam of $10^{-12}$ s order. The time scales for free electrons are therefore too short to generate non-trivial correlations to appear in the spectra at the observed frequency range. Second, the energy of the photons in the light beam are 2.4 eV which is of the same order as the work function for these metals of 4 5 eV. Therefore, observation of the fluctuations in the free electrons is expected to be non-passive and lead to nonlinear behavior. This is inconsistent with the linear behavior seen in the spectra, as in Fig. 6. Since the time scales are so short for free electron processes, these effects should not show up in the observed frequency range, though collective motions with the appropriate time scales might be able to explain the observed phenomenon.

Some of the possible sources of the observed fluctuations are the properties of the ion cores in the metal, in the light beam, down to the skin depth. The thermal motion of the cores, or photon interactions with the bound electrons can contribute to the observed fluctuations. The number density of ion cores is identical to that of the free electrons, up to the valency factor. The time scales of their thermal motion are much longer than the time scales of free electron motion, and the ion cores do not move out of the beam so that correlations arise within the observed time scales. It should be noted that the acoustic vibrations have long wavelengths compared to the separation of the beam locations, so that they do not contribute to the spectra. While the reflection is mainly caused by free electrons, their fluctuations are not observable in the correlations in the range of times that were observed here, so that the ion core contributions can dominate the observed fluctuation spectra. The spectra measured in this work have behaviors close to 1/f over a wide frequency range, and belong to a class often referred to as the “1/f noise”, which is widely seen in nature. In metals, thermal motion of atoms, including the effects of internal friction is known to lead to an 1/f spectrum, with additional frequency dependencies coming from the frequency dependency of the loss angle. These motions modulate the frequency of the light through Doppler shifts, which appear in the spectra, similarly to selective reflection. The loss angle values are not known with precision and the mechanism is technically involved. Considering photon interactions with bound electrons, whether they can produce photon correlations, as in, the linear response seen in Fig. and the shape of their spectra needs to be investigated.

When considering thermal fluctuations of any kind to be the source of the observed spectra, the temperature change due to light absorption needs to be considered. If the temperature increases significantly, the behavior becomes non-linear with respect to the light power, so that it is not compatible with the current observations. One can estimate the temperature rise of the sample beam spot roughly as follows: consider a uniform semi-sphere of radius $R$, with a semi-sphere with radius $w$ removed, at the center. When we dissipate heat from the inner sphere, the temperature difference $\Delta T$ between the inner and the outer boundary is $Q/(2\pi\kappa)(1/w - 1/R)$. Here, $Q$ is the heat dissipated from the inner surface and $\kappa$ is the thermal conductivity of the material. Considering our measurements, the absorptance of gold is almost an order of magnitude larger than that of aluminum or silver, while their thermal conductivities are similar, so that the gold surface may give rise to the largest temperature difference. For the maximal beam power in our experiments, the corresponding temperature rise is 2.7 K in the above simple formula. We have also computed the temperature rise numerically, with the experimental geometry, to find $\Delta T \approx 7.5$ K. This temperature rise, while larger than the result from the simple formula, is still much smaller than the room temperature ($\sim 300$ K), so that thermal fluctuations are quite compatible with being the source of the observed fluctuations.

More work needs to be done to clarify the dynamics behind the reflectance fluctuations we have observed, both
qualitatively and quantitatively. Measurements performed at different wavelengths can lead to more information, especially since the reflection mechanism depends on the wavelength of light\cite{2,4}. Fluctuations in the reflectance of metals yield another window into the mechanism underlying reflection, and understanding them would lead to deeper insight into the degrees of freedom contributing the reflection in metals. Similar measurements can be performed for different metals, other types of mirrors and various surfaces. How the spectrum depends on the material would be an intriguing question, and even more so, why.

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