Rapid Communications

Observation of quantum corrections to conductivity up to optical frequencies

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It is well known that the conductivity of disordered metals is suppressed in the limit of low frequencies and temperatures by quantum corrections. Although predicted by theory to exist up to much higher energies, such corrections have so far been experimentally proven only for $\lesssim$80 meV. Here, by a combination of transport and optical studies, we demonstrate that the quantum corrections are present in the strongly disordered conductor MoC up to at least ~4 eV, thereby extending the experimental window where such corrections were found by a factor of 50. The knowledge of both the real and imaginary parts of conductivity enables us to identify the microscopic parameters of the conduction electron fluid. We find that the conduction electron density of strongly disordered MoC is surprisingly high and we argue that this should be considered a generic property of metals on the verge of a disorder-induced localization transition.

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At finite frequencies ω, the optical conductivity of any material is a complex quantity, $\sigma(\omega) = \sigma'(\omega) + i\sigma''(\omega)$. In the limit of low frequencies, only the conduction band contributes to $\sigma'(\omega)$ of a metal; the conductivity of this band is customarily described by the Drude formula [1,2]

$$\sigma(\omega) = \frac{\sigma_0}{1 - i\omega/\Gamma}, \tag{1}$$

where $\Gamma = 1/\tau$ is the relaxation rate determined by the collision time of the electrons τ. Within the model of free electrons, $\sigma_0 = ne^2\tau/m$, where n and m are the electron concentration and mass, respectively [1].

With increasing disorder strength, Γ increases and σ_0 decreases, until at some critical disorder level the [three-dimensional (3D)] metal turns into an insulator [3] and $\sigma'(0)$ vanishes in the limit of low temperatures T. If Eq. (1) were to apply, $\sigma(\omega)$ would vanish identically for all ω. However, this is unphysical, since at nonzero frequencies the absorption has to be finite even in the insulating state. Therefore, in the insulating state, the low-frequency conductivity $\sigma'(\omega)$ has to grow with ω and, by continuity, the same behavior has to be expected also on the metallic side of the metal-insulator transition.

It is in fact well known that, in *weakly disordered* 3D metals, the conductivity in the small-ω and T limit can be described as $\sigma(\omega) = \sigma_{\rm reg}(\omega) + \delta\sigma(\omega)$, where $\sigma_{\rm reg}(\omega) \approx \sigma_0$ is the regular part of the conductivity and $\delta\sigma(\omega)$ is the quantum correction which grows with ω. Two mechanisms have been proposed for the latter: It can be either due to the so-called weak-localization corrections [4], or due to interaction effects [5]. Remarkably, up to numerical prefactors, at $T = 0$ both mechanisms yield the same functional form of the quantum correction [6] and its real part, $\delta\sigma'(\omega)$, can be written in a unified way as

$$\delta\sigma'(\omega) \approx \mathcal{Q}^2\sigma_0(-1 + \sqrt{\omega/\Gamma}). \tag{2}$$

The quantum correction is present only for $\omega \lesssim \Gamma$ and its magnitude is characterized by a dimensionless number $\mathcal{Q}$, to be called quantumness. We emphasize that our parametrization Eq. (2) reflects the fact that the quantum correction has to diminish the conductivity.

Numerical simulations by Weisse within the weak-localization scenario at $T = 0$ have shown that, for not too high ω, Eq. (2) is qualitatively valid not only for weakly disordered metals, but in a *broad range of disorder strengths* in the metallic phase [7]. Weisse's data indicate that $\mathcal{Q}$ is of the same order of magnitude as $\hbar\Gamma/\varepsilon_F$, where ε_F is the Fermi energy: In the limit of weak disorder this result is well known [6]; in the opposite limit when the metal-insulator transition is approached, the quantumness $\mathcal{Q} \to 1$ and $\hbar\Gamma$ is comparable to ε_F [8].

In order to generalize the formulas for $\sigma'(\omega)$ to finite T, we follow the recipes of Fermi-liquid theory and replace ω by $\varpi = \sqrt{\omega^2 + \gamma(T)^2}$, where $\gamma(T)$ is a T-dependent scattering rate which depends on the mechanism for the quantum corrections: In the case of weak-localization corrections it is the phase-breaking scattering rate, whereas for interaction effects $\gamma(T) = \pi k_B T/\hbar$. Detailed calculations [6] for weakly disordered metals do confirm such a procedure, again up to numerical prefactors.

Motivated by Weisse's data and analytical theory for weakly disordered metals, throughout the metallic phase we postulate the following simple formula for the ω and T dependence of the real part of the optical conductivity,

$$\sigma'(\omega, T) = \sigma_0[1 - \mathcal{Q}^2 + \mathcal{Q}^2\sqrt{\varpi/\Gamma}], \quad \text{if } \varpi < \omega^*, \tag{3}$$

$$\sigma'(\omega, T) = \frac{\sigma_0}{1 + (\varpi/\Gamma)^2}, \quad \text{if } \varpi \geqslant \omega^*. \tag{4}$$

This formula features the Drude behavior Eq. (1) in the high-frequency limit $\varpi \geqslant \omega^*$ and, at the same time, for $\varpi < \omega^*$ takes into account the 3D quantum correction Eq. (2). The

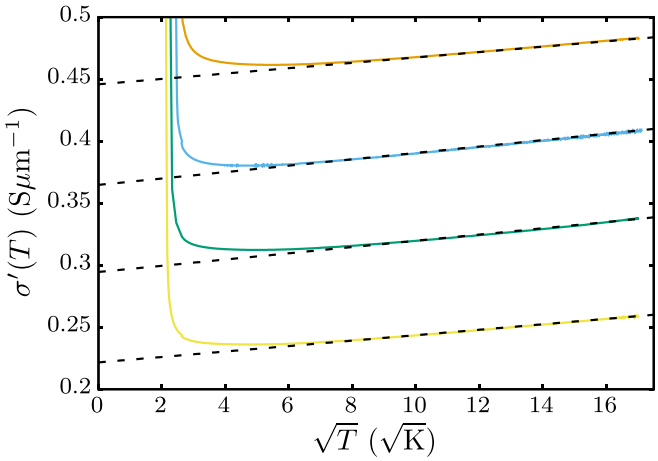

FIG. 1. Temperature dependence of dc conductivity for a series of 5-nm-thick MoC films. The sheet resistance of the films at room temperature is (from bottom to top) $R_\square = 720$, 590, 500, and 415 Ω. Fits to Eq. (3) are shown as well.

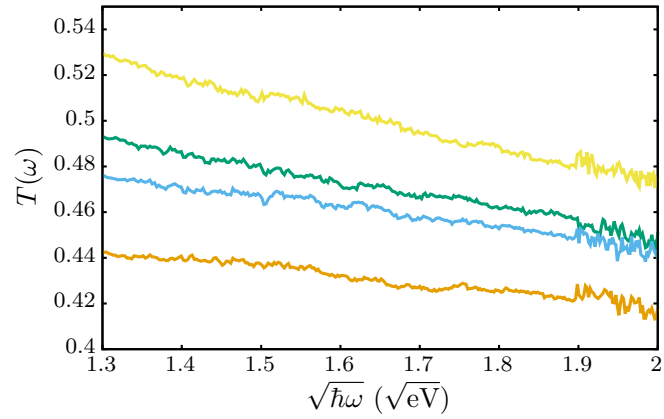

FIG. 2. Frequency dependence of transmission through MoC thin films on sapphire substrates for the same set of films and with the same color coding as in Fig. 1.

model Eqs. (3) and (4) depends on three parameters: σ_0, $\mathcal{Q}$, and Γ. Once these are known, the crossover frequency ω^* follows from assuming that the function $\sigma'(\omega, T)$ is continuous. We treat $\mathcal{Q}$ and Γ as independent, while for σ_0 we keep the expression $\sigma_0 = ne^2/(m\Gamma)$. Note that Eqs. (3) and (4) do not have to satisfy any partial sum rule for the conduction band [9].

Square-root corrections to conductivity in systems close to the metal-insulator transition have been previously observed in various materials. The best studied case is amorphous Nb:Si [10]. In metallic Nb:Si samples, very good agreement with a slightly modified version of Eq. (3) has been found [11] for $T \lesssim 16$ K and at frequencies up to 1 THz, both corresponding to $\hbar\varpi \lesssim 4$ meV.

The goal of this Rapid Communication is to check whether quantum corrections to conductivity are observable even at optical frequencies. To this end, we have chosen to study the highly disordered conductor MoC [12,13]. In this material quantum corrections to transport have been observed up to 300 K [14], corresponding to $\hbar\varpi \approx 80$ meV. The amount of disorder in MoC can be increased either by a reduction of the film thickness or by an increase of the carbon content.

In this Rapid Communication we study two sets of MoC films prepared on sapphire wafers by means of reactive magnetron deposition from a Mo target in an argon-acetylene atmosphere [15]. In the first set, we prepared films with thickness $d = 5$ nm and varying Mo:C stoichiometry, while in the second set we have studied films at fixed stoichiometry but with varying thickness [9].

The temperature dependence of dc conductivity $\sigma'(T)$ for the set of samples with fixed thickness is presented in Fig. 1. In a transport measurement $\omega \approx 0$, and therefore $\varpi = \gamma(T)$. As can be seen, $\sigma'(T)$ exhibits very good scaling with the square root of temperature from $T_{\min} \approx 50$ K up to room temperature, precisely as expected according to Eq. (3) in the case of dominant interaction effects with $\gamma(T) = \pi k_B T/\hbar$. The deviations from this scaling below $T_{\min}$ are caused by the superconducting transition and the associated fluctuation conductivity. Moreover, a dimensional crossover between 3D and 2D quantum corrections is expected to occur at temperatures comparable with $T_{\min}$ [9].

As can be seen from Fig. 1, the two terms in Eq. (3) exhibit a quite different evolution with stoichiometry: The extrapolated $\varpi = 0$ value of the conductivity $\sigma'(0)$ decreases when the metal-insulator transition is approached, whereas the coefficient in front of $\sqrt{T}$ is roughly constant. Similar behavior has been observed previously in Nb:Si [10,11] and in TiO_x [16]; it is also consistent with our model Eqs. (3) and (4) [9].

In Fig. 2 we show the optical transmission $\mathcal{T}(\omega)$ in a broad frequency range for the same set of films on sapphire substrates as in Fig. 1. The absence of any spectral features indicates that interband transitions are absent in this range, a point we will come to later. Similar featureless transmission data are also obtained for the set of films with varying thickness. This indicates that the details of the microstructure are not important for the phenomena we observe and that, for both sets of samples, the crucial control parameter is the degree of disorder [9].

The real part of the dimensionless sheet conductance [17] of the MoC films $g(\omega) = Z_0\sigma(\omega)d$, where Z_0 is the impedance of free space, can be calculated from the transmission $\mathcal{T}(\omega)$ [9]. The thus obtained conductance $g'(\omega)$ of a MoC film with thickness $d = 5$ nm and room-temperature sheet resistance $R_\square = 720$ Ω is shown in Fig. 3. Also shown in Fig. 3 is the ellipsometry data for both components of $g(\omega)$ which are obtained in a somewhat more narrow frequency range. Since at optical frequencies $\omega \gg \gamma(T)$, we do not have to distinguish between ω and ϖ. The temperature dependence of the conductivity for this sample is replotted here from Fig. 1 as well, assuming $\gamma(T) = \pi k_B T/\hbar$. Note the very good agreement between all three data sets.

Since conductivity σ and dimensionless conductance g differ only by a multiplicative constant, when talking about frequency and temperature dependence, from now on we will use these terms interchangeably.

The data presented in Fig. 3 are the main result of this Rapid Communication. It shows that the real part of conductivity of strongly disordered MoC thin films is very well

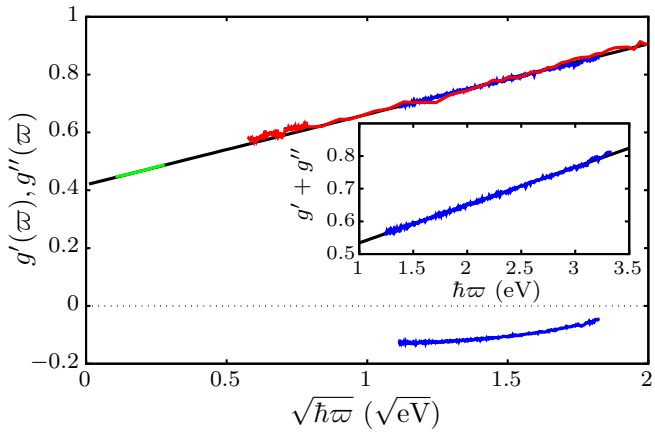

FIG. 3. Dimensionless sheet conductance $g(\varpi)$ of a 5-nm-thick MoC film with room-temperature sheet resistance $R_\square = 720\ \Omega$. Green: Data obtained from the temperature dependence of the dc conductivity in Fig. 1 assuming $\gamma(T) = \pi k_B T/\hbar$. Red: Data from optical transmission. Blue: Real (positive) and imaginary (negative) parts of $g(\varpi)$ determined by ellipsometry. Black line: Fit of the real part to Eq. (3). The inset shows that the anomalous terms proportional to $\sqrt{\varpi}$ perfectly cancel in $g'(\varpi) + g''(\varpi)$.

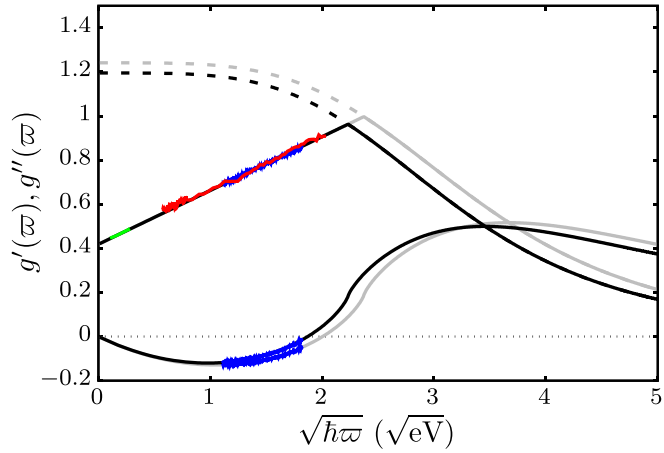

FIG. 4. Results of the prolongation procedure described in the text for $\epsilon_\infty = 1$ (gray lines) and $\epsilon_\infty = 1.4$ (black lines), applied to the data from Fig. 3 (with the same color coding). The dashed lines denote the continuation of the high-frequency behavior (i.e., conductance in the absence of quantum corrections) to low frequencies.

described by Eq. (3) in a broad range of frequencies from $\hbar\varpi \approx 14$ meV up to at least $\hbar\varpi \approx 4$ eV. Although this is expected from the theoretical point of view since the scattering rate Γ in dirty metals close to the metal-insulator transition is huge, we now demonstrate it experimentally. This fact is not generally adopted and it is often incorrectly assumed that quantum corrections are not present at room temperature or at optical frequencies.

The results for the imaginary part of conductivity $\sigma''(\omega)$ are also presented in Fig. 3. It should be pointed out that, unlike the real part of conductivity which is (in the studied frequency range) determined only by the contribution of the conduction band, there exists an additional contribution to $\sigma''(\omega)$ from the bound electrons, $\sigma''_{\text{bound}}(\omega) = -\epsilon_0(\epsilon_\infty - 1)\omega$, where ϵ_0 is the permittivity of vacuum and ϵ_∞ is the bound-electron contribution to the static dielectric constant. Thus the presence of a negative contribution to $\sigma''(\omega)$ is by itself not surprising. However, the experimental data in Fig. 3 clearly indicate that $\sigma''(\omega)$ is not linear in frequency. In fact, the data contain an anomalous term $-\mathcal{Q}^2\sigma_0\sqrt{\omega}$ with the same magnitude and opposite sign as in the real part Eq. (3). Such a term is required to be present by the Kramers-Kronig relations and in the inset to Fig. 3 we demonstrate that, as was to be expected, the anomalous terms perfectly cancel in the sum $\sigma'(\omega) + \sigma''(\omega)$.

The next natural question to ask is as follows: What are the values of the parameters σ_0, $\mathcal{Q}$, and Γ which enter Eq. (3)? From Fig. 3 we have access to only two parameters: the $\varpi = 0$ value of the conductivity $\sigma'(0)$ and the coefficient in front of $\sqrt{\varpi}$. On the other hand, if we could extend our measurements to higher frequencies and measure the crossover scale ω^* predicted by Eqs. (3) and (4), this would give us the needed third data point. Unfortunately, at frequencies above $\hbar\omega \sim 4$ eV the transmission of our sapphire substrates is influenced by impurity absorption and therefore we cannot measure ω^* directly.

Nevertheless, one can approximately determine the complex conductivity in the whole frequency range from a measurement of both $\sigma'(\omega)$ and $\sigma''(\omega)$ in a finite interval of frequencies [18]: Making use of the Kramers-Kronig relations, one can write down integral equations for the unknown conductivity outside the measured range. However, since the solution of these equations is numerically unstable, additional simplifying assumptions have to be made. We have used the following procedure for the prolongation of the $\sigma'(\omega)$ data, which turned out to be quite robust: We start by choosing a value of Γ. Having made this choice, we can unambiguously find σ_0 and $\mathcal{Q}$ from fitting the real part of conductivity to Eq. (3). With known σ_0, $\mathcal{Q}$, and Γ, the real part of conductivity is known from Eqs. (3) and (4) on the entire real axis. Next, we calculate, making use of the Kramers-Kronig relations, the imaginary part of the conductivity $\sigma''(\omega)$. Finally, we adjust the value of Γ so that good agreement with $\sigma''(\omega)$ of the conduction band is obtained.

Note that in order to obtain $\sigma''(\omega)$ of the conduction band, the contribution $\sigma''_{\text{bound}}(\omega)$, parametrized by ϵ_∞, should be subtracted from the ellipsometric data for the imaginary part of conductivity in Fig. 3. We estimate [9] that ϵ_∞ satisfies the bounds $1 < \epsilon_\infty < 1.4$.

The results of the described prolongation procedure for $\epsilon_\infty = 1$ and $\epsilon_\infty = 1.4$ are presented in Fig. 4. Both prolongations fit $\sigma''(\omega)$ of the conduction band very well and the variation between them is small. In order to further test the robustness of our prolongation procedure, we have modeled the real part of conductivity at high frequencies $\omega \geqslant \omega^*$ by the Gaussian formula $\sigma'(\omega) = \sigma_0 \exp(-\omega^2/\Gamma^2)$ instead of Eq. (4); the resulting fits of $\sigma''(\omega)$ are equally good as when Eq. (4) is used. We have also checked that smoothing the cusp at $\omega = \omega^*$ in the frequency dependence of $\sigma'(\omega)$ results in only marginal changes of the prolongations. Based on all of this evidence we conclude that our prolongation procedure is robust and the extracted parameters are [9] $\mathcal{Q} \approx 0.82 \pm 0.01$, $\hbar\Gamma \approx 11.85 \pm 1.75$ eV, and $g_0 \approx 1.26 \pm 0.06$, implying $n = (4.3 \pm 0.8) \times 10^{23}\ \text{cm}^{-3}$.

In what follows we will comment on the values of the extracted parameters. The large value of the quantumness $\mathcal{Q}$ implies that the $T = 0$ limit of the dc conductivity, $\sigma'(0) = (1 - \mathcal{Q}^2)\sigma_0$, is reduced from the naive value σ_0 roughly by a factor of 3. Therefore, if one were to interpret this limit of the measured dc conductivity as σ_0 (as is frequently done), one would overestimate the scattering rate Γ by the same factor of 3.

The extracted energy scale $\hbar\Gamma$ is surprisingly large. This does make sense, however: Up to 4 eV, the real part of conductivity is described by Eq. (3) without any noticeable higher powers of frequency. This must mean that the crossover scale $\hbar\omega^*$ is by a wide margin larger than 4 eV. If one further observes that for $\mathcal{Q} \approx 0.82$ we have $\hbar\omega^* \approx 0.5\hbar\Gamma$, the large value of $\hbar\Gamma$ seems to be inevitable.

Even more surprising is the very large extracted electron concentration n, which is more than twice as large as in metallic aluminum. We believe that this is a consequence of the large value of Γ: The individual electronic bands which are separated by energy less than $\hbar\Gamma$ lose their identity and merge together. In order to estimate the electron concentration predicted by such a picture, let us consider cubic MoC which crystallizes in the rocksalt structure with lattice constant 4.27 Å [19] and concentration of one type of atoms $n_{\mathrm{at}} = 5.1 \times 10^{22}$ cm^{-3}. The valence electron configurations of the Mo and C atoms are $4d^5\,5s^1$ and $2s^2\,2p^2$, respectively. According to band-structure calculations [19,20], the relevant $4d$ and $5s$ states of molybdenum as well as the $2s$ and $2p$ states of carbon are within $\pm\hbar\Gamma$ from the Fermi energy. Therefore it is reasonable to assume that the conduction electron fluid is formed by all ten valence electrons and the corresponding electron density is $n = 10 \times n_{\mathrm{at}} = 5.1 \times 10^{23}$ cm^{-3}, a value within the error bar of the extracted n. As a matter of fact, the value of n_{at} in a highly disordered material is expected to be lower than the value for a perfect crystal which we have used; this (as well as an excess of carbon atoms which we also did not take into account) should improve the agreement between the estimated and extracted values of n. It is also worth pointing out that the absence of interband transitions up to 4 eV (see Fig. 2) provides an additional nontrivial consistency check of our proposal that the conduction electron "band" is very broad.

We believe that the electron concentration n in the conduction fluid of a dirty metal close to a disorder-induced metal-insulator transition should be generically large. In fact, e.g., in dirty NbN samples similarly large values of n have already been observed [9,21].

It is worth pointing out that the large value of n implies a large Fermi energy ε_F: Making use of the free-electron estimate $\varepsilon_F = \hbar^2(3\pi^2 n)^{2/3}/(2m)$ we find $\varepsilon_F = 20.65 \pm 2.55$ eV. Combining this with the knowledge of Γ, for the dimensionless parameter $k_F\ell = 2\varepsilon_F/(\hbar\Gamma)$ we obtain $k_F\ell \approx 3.5 \pm 0.1$. A nontrivial check of this analysis can be performed as follows. From a comparison of the conventional formulas for the quantum corrections [6] to our Eq. (3), it follows that the product $\mathcal{Q} \times k_F\ell$ should be of the order 1. This constraint turns out to be well satisfied by our data, since $\mathcal{Q} \times k_F\ell \approx 2.9$.

In conclusion, we have shown that, in strongly disordered metals on the verge of a disorder-induced localization transition, quantum corrections to conductivity may be present up to optical frequencies. This effect should be universal; therefore quantum corrections should be added to the list of known reasons [2] why the canonical Drude formula for the frequency dependence of conductivity is hard to observe. We speculate that quantum corrections at infrared frequencies and above may already have been observed previously, but they were interpreted in a different way. Most notable candidates are liquid mercury [22] and perhaps also Si:P [23]. It therefore seems worthwhile to also take the quantum corrections into account in models used for spectroscopic ellipsometry.

We have likewise demonstrated how the combined knowledge of both the real and imaginary parts of the optical conductivity can be used to extract microscopic parameters of the conduction electron fluid in dirty metals, such as the quantumness $\mathcal{Q}$, the scattering rate Γ, and the electron concentration n. In particular, the magnitude of the quantum correction $\delta\sigma' = -\mathcal{Q}^2\sigma_0$ in the low-T and ω limit is currently not accessible in any other way. We have found that n is very large in MoC close to the metal-insulator transition; its value indicates that the conduction electron fluid is formed by all valence electrons. We have argued that the large value of n should be a generic property of dirty metals, since individual electronic bands which are separated by energy less than $\hbar\Gamma$ lose their identity and merge together.

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