Optical Properties of Ultrathin Films: Evidence for a Dielectric Anomaly at the Insulator-to-Metal Transition

J. J. Tu,* C. C. Homes, and M. Strongin

Department of Physics, Brookhaven National Laboratory, Upton, New York 11973-5000

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Optical properties of ultrathin layers (< 50 Å) of Au and Pb quench condensed on amorphous germanium (α-Ge) have been measured *in situ* at 10 K. The development of these films from an insulating state to a metallic state is traced as a function of the film thickness as well as the sheet resistance, $R_\square$. Of particular interest is the regime of $R_\square$ near 3000 Ω where there is an anomaly in the optical transmission. This anomaly is due to a singularity in the dielectric function when the system undergoes an electronic percolation or insulator-to-metal (I/M) transition.

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The insulator-to-metal (I/M) and insulator-to-superconductor (I/S) transitions are fundamental issues in modern condensed matter physics. Questions even remain as to whether there exists a true metallic state at $T = 0$ in two-dimensional systems [1]. Ultrathin metal films (UTMF) quench-condensed on α-Ge at helium temperatures [2] have served as model systems for the study of I/S and I/M transitions in 2D (e.g., see the review [3]) and more recently have been shown as a paradigm for high T_c cuprates [4]. While these films had been thought to be "homogeneous" [2,5,6] in contrast to granular films made without α-Ge, recent *in situ* STM studies have revealed structures on the nanoscale [7] that can profoundly influence their electronic properties. Traditionally, the I/S and I/M transitions in UTMF have been examined using the temperature gradient of the resistivity, $d\rho/dT$ [3,8]: The I/S transitions have been found to occur at the nearly universal value of $\sim h/4e^2 = 6450\ \Omega$ for superconducting UTMF, but no I/M transitions have been unambiguously identified in non-superconducting UTMF. However, the dc transport measurements have some limitations: (i) Measurements can be done only in a limited temperature range (< 15 K) in order to fix the film morphology; (ii) while the electronic properties of UTMF are determined by their complex conductivity, only the dc values can be determined. Hence, by measuring the frequency dependent optical transmission of both types of UTMF, most of these limitations can be avoided to shed more light on the I/S and I/M transitions in these systems.

In this Letter, the development of ultrathin Au and Pb films from an insulating state to a metallic state is examined at a fixed temperature of 10 K by measuring the far-infrared transmission as a function of film thickness. An anomaly in the optical transmission is observed for films with $R_\square$ of $\sim 3000\ \Omega$ (thickness ~ 20 Å) due to an electronic percolation or I/M transition. It should be noted that this transition occurs at a smaller resistance value than the nearly universal value of $R_\square \sim h/4e^2 = 6450\ \Omega$ for the I/S transition.

While the optical conductivity of thick and granular films has been investigated extensively [9], there has been only one study on ultrathin films [10] and none for UTMF in the far-infrared region. For this study, ultrathin layers (< 50 Å) of Au and Pb are quench condensed on (111)-cut silicon substrates with an α-Ge underlayer (~ 10 Å) in ultrahigh vacuum ($\sim 5 \times 10^{-9}$ Torr) at 10 K: the same method that has been used to make UTMF since about 1970. The dc sheet resistance, $R_\square$, is monitored by a standard four-probe technique and the far-infrared transmission is measured *in situ* on a Bruker IFS 113v spectrometer.

In Fig. 1, the raw transmission data is presented for both Pb and Au films at 10 K. The transmittance $T(\omega)$ (for a thin film deposited on a substrate) is defined as a ratio in the usual fashion [9]: $T(\omega) = T_{\text{substrate+film}}/T_{\text{substrate}}$, where $T_{\text{substrate+film}}$ and $T_{\text{substrate}}$ are the transmittance of the substrate with and without the film, respectively. For metallic thin films, this ratio is given by the so-called Tinkham formula [11]:

$$T(\omega) = \frac{1}{|1 + \tilde{\sigma} d \frac{Z_0}{n+1}|^2}, \tag{1}$$

where $Z_0 = 377\ \Omega$ is the impedance of free space, n is the index of refraction of the substrate, d is the thickness of the film, and $\tilde{\sigma} d$ is referred to as the sheet conductance. Equation (1) is a good approximation as long as d is small and the interference effects within the film can be neglected [12]. The condition for this to hold is that the optical path length of the film is small so that there is no significant phase shift at the film/substrate interface. Obviously for UTMF, Eq. (1) is expected to hold because d is typically many orders of magnitude smaller than the wavelength of the far-infrared radiation. As shown in Figs. 1(a) and 1(b), the transmittance decreases with increasing film thickness as can be described by the Tinkham formula for the most part. However, anomalous behavior is observed between 15–21 Å for Au and 19–35 Å for Pb that is reproducible for the same thickness and resistance intervals.

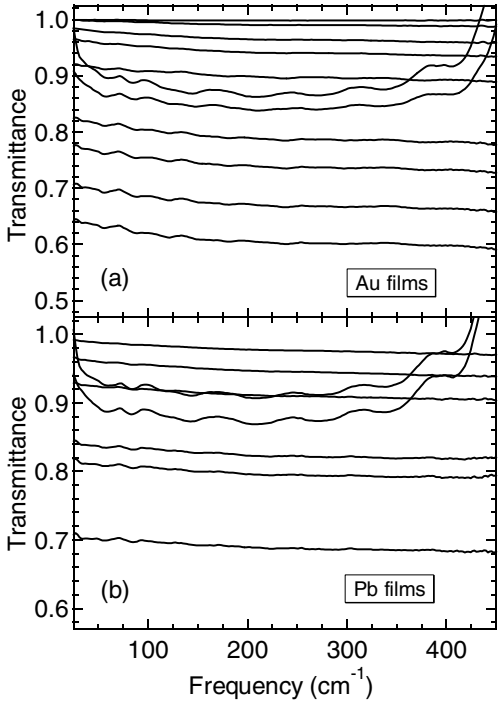

FIG. 1. Far-infrared transmission of UTMF at 10 K. The different curves correspond to different thicknesses. (a) Au films, from top down: 3, 6, 9, 12, 15, 21, 24, 30, and 35 Å. The two curves with anomalous frequency dependence correspond to thicknesses of 18 and 19 Å, respectively. (b) Pb films, from top down: 13, 16, 19, 35, 40, and 51 Å. The two curves with anomalous frequency dependence correspond to thicknesses of 21 and 28 Å, respectively.

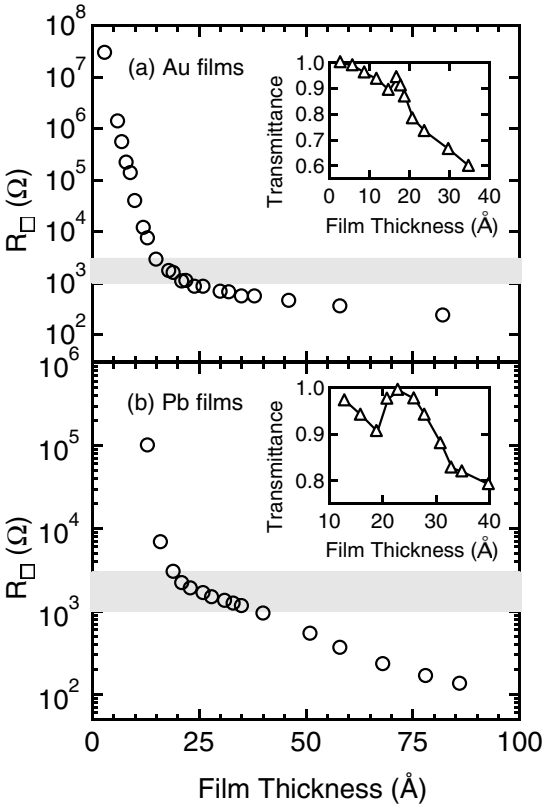

FIG. 2. The thickness dependence of the sheet resistance, $R_\square$, of UTMF. (a) Au films; (b) Pb films. The shaded section corresponds the region where the anomalous transmission occurs. Insets: The thickness dependence of the transmittance at a fixed frequency of 425 cm^{-1}.

Interestingly, in Fig. 2 there is no corresponding critical behavior in $R_\square$ between ~3000–1000 Ω where the anomalous transmission occurs for both systems. This should be compared with the thickness dependences of the transmission at a fixed frequency of 425 cm^{-1} that are given in the insets of Fig. 2 where a discontinuity in transmittance can be clearly identified in the anomalous region. These semilog plots, of $R_\square$ versus d, show that $R \propto \exp(-\alpha d)$. Even though there is no discontinuity in $R_\square$, the exponent α changes significantly before and after the transition region. The observation of such a change in $R_\square$ indicates that the region of anomalous transmission separates two electronic states of UTMF.

To understand the implications of Figs. 1 and 2, some discussions of the anomalous region are in order. The optical transmission through UTMF is completely determined by their complex dielectric function ($\tilde{\epsilon} = \epsilon_1 + i\epsilon_2$) or their complex conductivity ($\tilde{\sigma} = \sigma_1 + i\sigma_2$) that are related in the following way: $\tilde{\epsilon} - \epsilon_\infty = (4\pi i\tilde{\sigma})/\omega$. In the far-infrared region, ϵ_∞ can be neglected so that $\sigma_1 = \omega\epsilon_2/4\pi$, $\sigma_2 = -\omega\epsilon_1/4\pi$, and $1/R_\square = \sigma_\square(\omega = 0) = d\sigma_1(\omega = 0)$. The transmittance in the region of $R_\square \sim$ 3000–1000 Ω is anomalous mainly in two ways: (i) The oscillations in the transmittance correspond to exactly the oscillations that are present in the raw power spectrum (caused by phonon absorptions in the beam splitter and the cryostat windows) that are normally canceled out in the ratio $T(\omega)$; (ii) the transmittance is higher than the expected value as shown in the insets of Fig. 2 which means that the films in the anomalous region are acting effectively as antireflection coatings. These two observations imply that there is an anomaly in the complex dielectric functions that causes the Tinkham formula to break down in this region. However, there is no discontinuity in $R_\square$ which means there is no anomaly in σ_1 or ϵ_2. Hence, we argue that there must be an anomaly in σ_2 or ϵ_1. Furthermore, an anomaly in ϵ_1 occurs naturally at an electronic percolation transition or I/M transition where ϵ_1 diverges. This was termed the "dielectric catastrophe" by Mott [13] or at the Anderson I/M transition [14] due to disorder. In this light, near the I/M transition where ϵ_1 tends to diverge, the optical thickness of the film ($n_f d$), where n_f is the film index of refraction, can become comparable to the wavelength of the light (~0.01 cm). Therefore, the interference within the film can no longer be neglected, which means the Eq. (1) is not valid near the I/M transition. As a result, the oscillations start to appear in the ratio as well as the antireflection effects. It

is interesting to note that, in the transition region, the $R_\square$ values of ~3000–1000 Ω combined with the corresponding film thicknesses ($\rho = dR_\square$) give the resistivity values, 600–200 $\mu\Omega$-cm, that fall exactly into the so-called Ioffe-Regel limit [15] which is considered as the natural divide between insulating and metallic states.

Away from the transition region, the optical spectra of these UTMF in the insulating and metallic states are well behaved. In these regimes, the Tinkham formula will be further approximated: The contribution of the imaginary part of conductance to the transmittance can be neglected [10], so that $\sigma_\square(\omega)$ can be determined from the transmission data:

$$\sigma_\square(\omega) = \frac{n+1}{Z_0}\left(\sqrt{\frac{1}{T(\omega)}} - 1\right). \quad (2)$$

In Fig. 3, $\sigma(\omega)$ for both Au and Pb films are shown in a log-log plot. The slopes of the linear fits to these spectra give the exponent β in the power law relation: $\sigma_\square(\omega) \propto \omega^\beta$. The properties of the films are different in the insulating region as compared to the metallic region because the exponent β exhibits distinct behaviors. In the insulating region, β changes rapidly with film thickness, while in the metallic region β has practically the same value. The power law behavior in the insulating state has been predicted by models relying on ac hopping of pairs of states [16]. It should be noted that β has a value of 0.9 for the thinnest Au film of 6 Å in Fig. 3, and this value is quite close to the value that is derived from these models when Coulomb interactions are included [16,17].

In Fig. 4, $\sigma_\square(\omega)$ for both Au and Pb films are presented instead in a semilog plot. In the metallic region, the slopes γ of these $\sigma_\square(\omega)$ vs $\ln\omega$ curves can be compared with the prediction of 2D weak localization theory [18] that gives a universal value of $\gamma = e^2/h\pi = 1.24 \times 10^{-5}\ \Omega^{-1}$. The slopes γ show different trends in the insulating and metallic regions in that γ tends to saturate in the metallic region particularly for Pb but at a value that is about 7 times that of the universal value. Therefore, the frequency dependence of $\sigma_\square(\omega)$ for these UTMF cannot be described by the 2D weak localization theory even if the interactions between the electrons are considered. The much stronger frequency dependence of $\sigma_\square(\omega)$ implies that UTMF are not uniform 2D electronic systems and that the nanoscale inhomogeneity in UTMF is essential to any theoretical descriptions [10].

The important finding is the observation of a dielectric anomaly near $R_\square \sim 3000\ \Omega$ in UTMF due to a percolative I/M transition. The thickness dependence of $R_\square$ also reenforces this conclusion. In fact, previous photoemission measurements lend some support for precisely such an I/M transition [19]. They show that for Pb films deposited on α-Ge at 10 K, the Ge band gap decreases with metal deposition. The electronic states approach the Fermi level at about 15 Å which is consistent with the

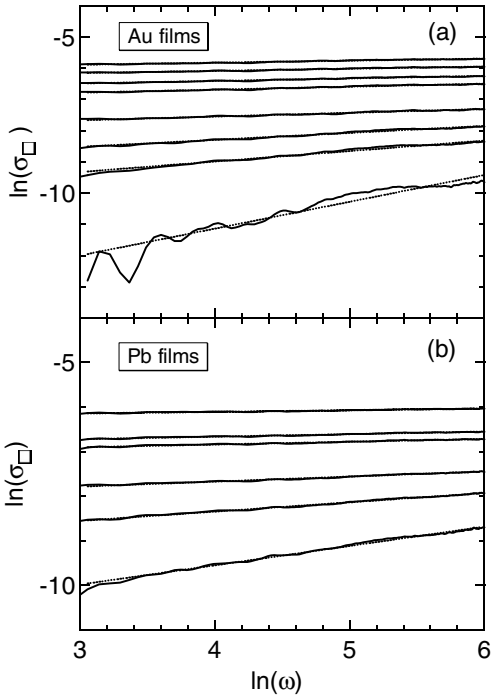

FIG. 3. Sheet conductance of UTMF at 10 K in a log-log plot. The different curves correspond to different thicknesses, and values of the slope β are given in the bracket. (a) Au films, from bottom up: 6 Å(0.9), 9 Å(0.3), 12 Å(0.2), 15 Å(0.1), 21 Å(0.09), 24 Å(0.08), 30 Å(0.07), and 35 Å(0.07); (b) Pb films, from bottom up: 13 Å(0.4), 16 Å(0.2), 19 Å(0.1), 35 Å(0.06), 40 Å(0.06), 51 Å(0.04).

current observation of a dielectric anomaly at ~20 Å. It is interesting that recent STM studies of quench-condensed Au films clearly show a qualitative change in film morphology at ~20 Å [7], and this coincides with the electronic I/M transition observed here. Also, an I/M transition at ~20 Å in these UTMF has been predicted by Phillips *et al.* [20], and he argues that the I/M transition and the structural change should take place at the same place due to the dynamic phonon blockade.

The idea of nanoscale inhomogeneity provides a picture of why the I/M transition occurs at a smaller sheet resistance $R_\square \sim 3000\ \Omega$ compared to the nearly universal value of $\sim h/4e^2 = 6450\ \Omega$ [21] for the I/S transition in UTMF (e.g., for Pb films [22] and Bi films [23]). The I/M transition discussed here is indeed a different transition from the I/S transition when the disorder in the films is varied because, below 15 K, the sheet resistance of UTMF with $R_\square < 6500\ \Omega$ is practically a constant of temperature. At initial stages of UTMF growth, nanoscale metallic regions, in, e.g., Pb films, are formed but are disconnected from one another. In this insulating state, their size is small enough so that, for temperatures below the bulk T_c, k_BT is greater than the condensation energy, $VH_c^2/8\pi$, and order parameter fluctuations (both amplitude and phase) prevent a global superconducting state from forming. However, when $R_\square$ becomes $\sim h/4e^2$, the

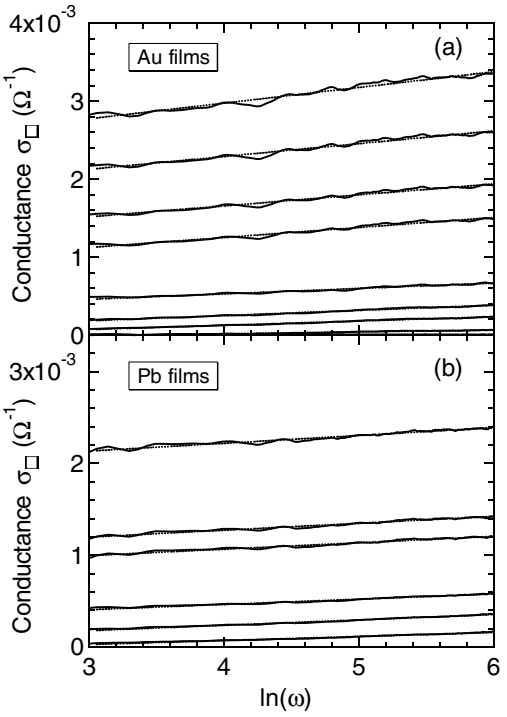

FIG. 4. Sheet conductance of UTMF at 10 K in a semilog plot. The different curves correspond to different thicknesses, and values of the slope γ are given in the bracket. (a) Au films, from bottom up: 6 Å(3×10^{-5}), 9 Å(5×10^{-5}), 12 Å(6×10^{-5}), 15 Å(7×10^{-5}), 21 Å(1×10^{-4}), 24 Å(1×10^{-4}), 30 Å(2×10^{-4}), and 35 Å(2×10^{-4}); (b) Pb films, from bottom up: 13 Å(4×10^{-5}), 16 Å(5×10^{-5}), 19 Å(6×10^{-5}), 35 Å(7×10^{-5}), 40 Å(8×10^{-5}), and 51 Å(8×10^{-5}).

screening is adequate to reduce the Coulomb energy and Josephson tunneling occurs between the nanoscale metallic regions, and the system lowers its free energy by forming a superconducting state. Eventually, as more metal is deposited, $R_\square$ is lowered further and the percolative I/M transition can be observed when superconductivity is removed by applying a magnetic field [24] or by staying in the normal state (as in the present case). From this quantum percolation point of view, the value of $R_\square$ at which the I/M transition occurs should depend on the morphology of UTMF and does not have a universal value. In this regard, UTMF can be considered as model systems for high T_c cuprates [4] particularly in the underdoped region (due to the percolative nature) to address the issues such as the pseudogap and the non-Drude-like optical conductivity. Whether the I/M and I/S transitions both occur near the maximum value of $R_\square \sim h/e^2 = 26\ \mathrm{K}\Omega$ for uniform 2D electron systems [25] should be further investigated.

In summary, an anomaly in the optical transmission is observed in UTMF due to an electronic percolation or I/M transition. Perhaps the most important finding is that as a function of disorder the I/M transition occurs at a lower resistance (less disorder) than the I/S transition for these systems.

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*Electronic address: jtu@bnl.gov

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