Terahertz conductivity of thin metal films

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The conductivities of thin Al, Au, and Ag films were measured via their transmission at terahertz frequencies. The conductivities of all the films, particularly the thinner films and Al films, were much smaller than their bulk dc values. This reduced conductivity can be quantitatively understood in terms of an increased scattering rate from defects. The transmission is consistent with a frequency independent conductivity, implying a very fast electron scattering time. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2968308]

The conductivity of metals at terahertz (THz) frequencies is becoming increasingly important as both microcircuit technology and microwave resonators are being driven at ever-higher frequencies. Our earlier work with terahertz metal parallel-plate waveguides[1] has shown that the conductivity of Al and Cu at THz frequencies is considerably less than the expected handbook value, particularly at cryogenic temperatures, due to defects within the 100 nm skin-depth layer. We have now extended and confirmed these results by measuring the terahertz transmission of a number of thin films comprised of different metals and different thicknesses. Unlike our previous work, this allows us to measure the conductivity of the thin film as a function of the thickness, since we are probing the entire film and not just the skin-depth layer.

Thin films of Al, Au, and Ag were deposited on high-resistivity, 2 in. diameter, 0.43 mm thick Si wafers via thermal evaporation. The typical evaporation rates were 2 nm/s onto an unheated wafer in order to ensure a relatively high conductivity film. The thickness of the film was measured via a quartz thickness monitor. Six films were made in total: Al films of 36, 88, and 152 nm thickness, Au films of 85 and 150 nm thickness, and an Ag film of 86 nm thickness. The films were not annealed after evaporation. The metal coated wafers were placed in a standard terahertz time-domain spectroscopy (THz-TDS) apparatus.[2,3] Their transmission was measured at both 295 and 77 K. The terahertz pulses and their amplitude spectra are shown in Fig. 1.

The amplitude transmission at a frequency f of the films and wafers can be modeled by the thin film formula[4]

$$t = \frac{t_{12}t_{23}t_{34}\exp[i(2\pi hf/c)n_2]\exp[i(2\pi df/c)n_3]}{1 + r_{12}r_{23}\exp[2i(2\pi hf/c)n_2]}, \qquad (1)$$

where t_{ij} and r_{ij} are the complex Fresnel transmission and reflection coefficients,[4] respectively, medium 1 and 4 are vacuum, medium 3 is Si with a thickness $d=0.43$ mm and with a THz frequency independent index $n_3=3.4175$ and negligible terahertz absorption,[5] and medium 2 is the metal film with a thickness of h and a complex index[4,6] of

$$n_2 = (1+i)\sqrt{\sigma/(4\pi\varepsilon_0 f)}. \qquad (2)$$

Propagation within the metal is described by $\exp[ikz-\omega t]$ where $k=(2\pi f/c)n_2$. Using Eq. (2), one obtains the amplitude absorption coefficient α to be $\alpha=\mathrm{Im}(k)=1/\delta=\sqrt{\pi f\sigma\mu_0}$. Note that α is simply $1/\delta$, where δ is the usual result for the skin depth.[6,7]

For our case of a 88 nm Al film with a conductivity of $\sigma=16(\mu\Omega)^{-1}$, $n_2=(1+i)370$ at 1 THz, the resulting amplitude attenuation traversing a single pass within the metal film [i.e., $\exp(-\alpha z)$] is only 49%, illustrating that the low transmission of these films is dominated by the high reflections from surfaces 1,2 and 2,3. Given the high reflectivity of these metal films, the amplitude transmission t tended to be low, on the order of 10^{-2}–10^{-3}, corresponding to a power transmission on the order of 10^{-4}– 10^{-6}.

The measured amplitude transmissions t of the 88 nm Al film, the 85 nm Au film, and the 86 nm Ag film are shown in Figs. 2–4, along with the theoretical curves [from Eqs. (1) and (2)] and for which the frequency independent conductivity was adjusted for the best fit. The fitted conductivities are shown in Table I. The conductivity of all the films is considerably less than the handbook[8] bulk dc value. In general, the conductivity is less for thinner films, particularly at 77 K. Films with higher percentage conductivity relative to the bulk conductivity at 295 K showed a larger relative conductivity increase at lower temperature. This would be consistent with the films having additional, temperature independent electron scattering mechanisms compared to the bulk,

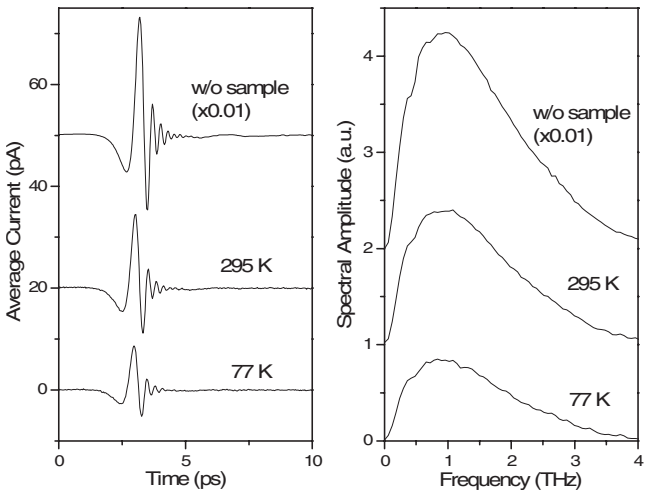

FIG. 1. Terahertz pulses through 88 nm Al film and corresponding spectra. Signals at 295 K and reference signal (without sample) are offset for clarity. Reference signal is multiplied by a factor of 0.01.

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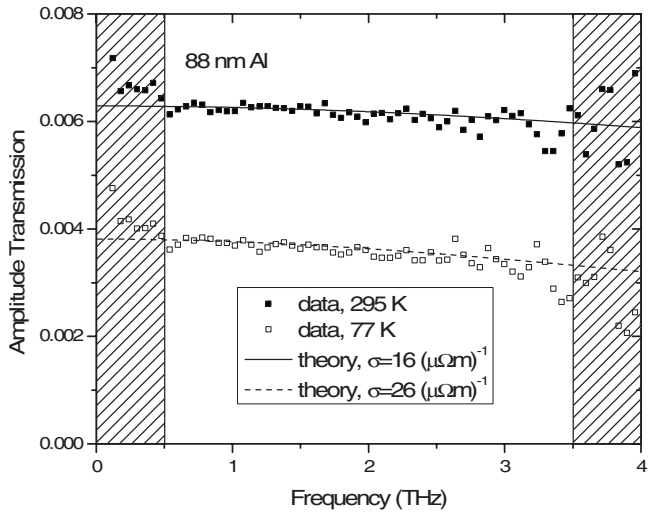

FIG. 2. Amplitude transmission t through 88 nm Al film at 295 and 77 K. The shaded regions correspond to frequencies with lower signal to noise.

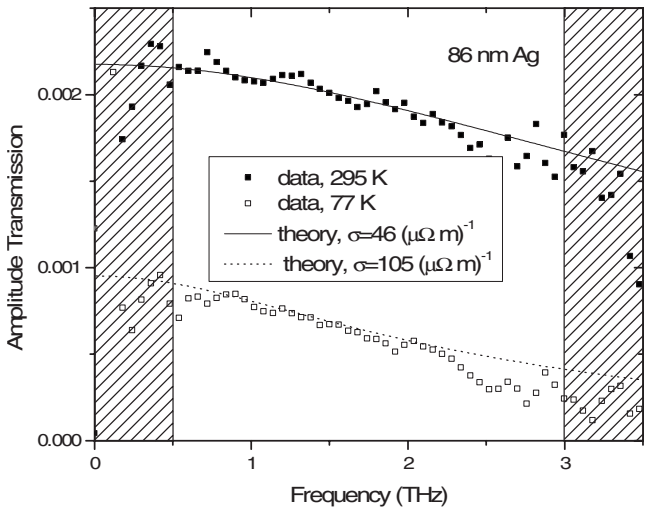

FIG. 4. Amplitude transmission t through 86 nm Ag film at 295 and 77 K.

such as defects, grain boundaries, and surfaces. These mechanisms would be more prevalent in the thinner films, resulting in a lower relative conductivity.

There is also a marked difference between the different metals. The Al films had the lowest conductivity compared to the bulk values (42%–58% at 295 K and 6%–12% at 77 K) while the Au and Ag films had the highest (33%–73% at 295 K and 12%–34% at 77 K). The Ag film (86 nm) had the additional distinction of having similar behavior to the thicker Au film at 150 nm. This would imply that the quality is best for the Ag film and worst for the Al films.

The dependence of the conductivity of thin metal films at dc frequencies has been well studied by the four-point probe method.[9,10] The dc conductivity of thin films is considerably less than the bulk conductivity and is reduced as the thickness of the film is reduced. The general consensus is that this low conductivity is due to the scattering from grain boundaries.[9,10]

For Al and Au, the conductivity measured with the dc thin film experiments at 80 K is two to four times greater than the corresponding conductivity measured with terahertz. For Ag, the conductivity measured by the dc thin film experiments at 80 K is only 10% greater than the corresponding terahertz measurements. This large difference between the dc and terahertz measurements may be due to different conductivities near different interfaces of the metal film. Our earlier work[1] revealed that the metal near the metal/air interface has considerably smaller conductivity than the metal near the metal/Si interface. The metal in the interior of the film would be expected to have the least number of defects, and hence the highest conductivity. While the dc work samples the entire thickness in a relatively uniform manner, our terahertz measurements are particularly sensitive to the conductivity near the interfaces, as they contribute to the very large reflections.

The thicknesses of all of the films measured in this paper were all larger than the percolation transition.[11] In addition, the film thicknesses are all larger than the mean free path of the electrons given either the measured conductivities or the bulk conductivities at 295 K.[12]

For the thinner films with less conductivity, the losses in the metal film itself have only slight positive frequency dependence (i.e., the higher frequencies have a smaller skin depth, corresponding to larger losses). This is offset by the inverse frequency dependence of the losses due to the reflections, resulting in a nearly frequency independent transmission. For the thicker films with more conductivity, the frequency dependence of the losses in the metal film dominate, resulting in a smaller transmission at higher frequencies.

In general, the conductivity can also be frequency dependent. This is most simply modeled by the Drude

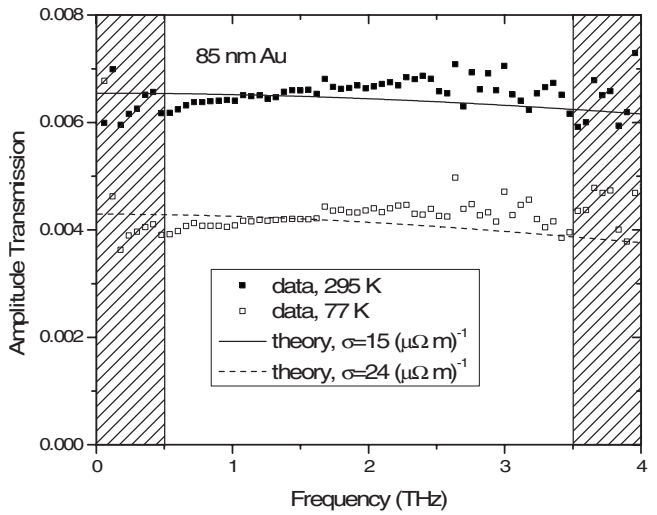

FIG. 3. Amplitude transmission t through 85 nm Au film at 295 and 77 K.

TABLE I. Conductivity $[(\mu\Omega\ \mathrm{m})^{-1}]$ of films; indicated percentage is relative to bulk conductivity from Ref. 8.

Metal	Thickness (nm)	σ at 295 K	σ at 77 K	Bulk σ_{dc} [a] at 295 K	Bulk σ_{dc} [a] at 80 K
	36	17 (45%)	25 (6%)		
	88	16 (42%)	26 (6%)		
Al	152	22 (58%)	51 (12%)	37.7	408
	85	15 (33%)	24 (12%)		
Au	150	31 (69%)	70 (34%)	45.2	208
Ag	86	46 (73%)	105 (30%)	63.0	346

[a]Reference 8.

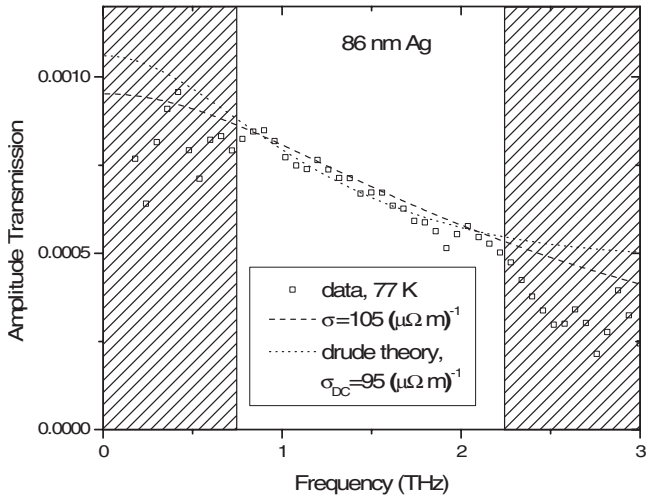

FIG. 5. Amplitude transmission t through 86 nm Ag film at 77 K, with different models: frequency independent conductivity and Drude model.

formula $\sigma = i\varepsilon_0\omega_p^2/(2\pi f + i\Gamma) = \sigma_{dc}(i\Gamma)/(2\pi f + i\Gamma)$, where $\sigma_{dc} = \varepsilon_0\omega_p^2/\Gamma$ is the dc conductivity and Γ is the electron scattering rate. The conductivity is reduced as the frequency f approaches the scattering rate $\Gamma/2\pi$. One can estimate $\Gamma/2\pi$ by scaling the bulk value[12] inversely by the same ratio of the measured σ to the bulk σ. These estimated values of $\Gamma/2\pi$ for the Al films range from 15 to 46 THz, resulting in only a very small difference from the frequency independent conductivity. For the Au films, the estimated values of $\Gamma/2\pi$ range from 3.4 to 16 THz, resulting in a minor difference in the shape of the frequency dependent transmission.

However, for the Ag film conductivity at 77 K, $\Gamma/2\pi = 2.4$ THz. This results in a predicted reduction in the conductivity at higher frequencies, resulting in a larger transmission. The measured transmission of the 86 nm Ag film at 77 K is compared in Fig. 5 to the predicted transmission given both a model with a frequency independent conductivity and the Drude model. Despite the low random error, there is a strong possibility of systematic error for transmission amplitudes below 0.0005. Consequently, we cannot distinguish between a frequency independent conductivity and the Drude model, which both give acceptable fits between 0.75 and 2.25 THz.

For scattering rates that are much higher than the considered frequency, the Drude model conductivity is essentially constant and given by $\sigma_{dc} = \varepsilon_0\omega_p^2/\Gamma$. According to Matthiessen's rule, the total scattering $\Gamma = \Gamma_P + \Gamma_D$ is the sum of a temperature dependent term Γ_P, dominated by phonons, and a temperature independent term Γ_D, dominated by defects such as grain boundaries. Typically, the scattering of high quality bulk material is dominated by Γ_P at room temperature. Upon cooling, Γ_P can be reduced until the scattering is dominated by Γ_D. For lower quality materials, Γ_D can be a strong component of scattering even at room temperature. For example, the scattering of bulk Al at 295 K is $\Gamma_P/2\pi \sim \Gamma/2\pi = 20$ THz.[12] However, the scattering of the 88 nm thin film at 77 K as estimated from the measured conductivity is $\Gamma_D/2\pi \sim \Gamma/2\pi = 29$ THz. Assuming Γ_P for the film is the same as for bulk, the scattering of the film at 295 K should be $\Gamma/2\pi = \Gamma_P/2\pi + \Gamma_D/2\pi = 49$ THz. This agrees reasonably well with the estimated Γ from the measured 295 K conductivity of $\Gamma/2\pi = 46$ THz. Reasonably good agreement is also obtained for the Au and Ag films. In general, the films with the lower relative conductivities have the highest Γ_D, resulting in only a minor change upon cooling. In contrast, the films with the higher relative conductivities generally have the lowest Γ_D, resulting in a sizable change upon cooling.

In summary, the essentially frequency independent conductivities of Al, Au, and Ag thin films have been measured via their THz transmission from 0.5 to 3 THz, and found to be much less than their bulk values, particularly at cryogenic temperatures. Our results are consistent with additional scattering mechanisms such as grain boundaries being more important with thin films. These results strongly impact electronic design and metallic cavities and waveguides at these frequencies. The Ag film had much higher conductivity than Al and Au films of the same thickness, which may be technologically important. These results are of particular importance to research studies with thin metal film THz surface plasmons[13–15] and metamaterials.[16]

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