Terahertz Spectroscopy of Ultrafast Carrier Dynamics in Nanomaterials

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***Abstract*—We use time-resolved terahertz spectroscopy to explore ultrafast carrier dynamics, transport, and localization in photoexcited nanocrystalline silicon films. The measured terahertz conductivity reveals a transition from free to localized behavior as the amount of disorder in the film increases, which is well-described within the framework of the Drude-Smith model.**

I. Introduction And Background

Terahertz pulses are ideal for probing ultrafast carrier dynamics and the nature of conduction in materials [1,2]. Many studies using terahertz time-domain spectroscopy (THz-TDS) or time-resolved THz spectroscopy (TRTS) techniques have shown that the terahertz conductivity of carriers in bulk semiconductors is reasonably described by the Drude model [3]. In nanomaterials, however, significant carrier localization effects lead to distinct non-Drude behavior in the observed terahertz conductivity, such as a suppression of the real part of the conductivity at low frequencies. Various conductivity models have been used to describe this behavior, including effective medium [4], polarization [5], and plasmonic models [6]. A generalized version of the Drude model developed relatively recently by Smith [7] has also been used to describe the terahertz conductivity observed in TiO_2 [8] nanoparticles, nanostructured ZnO films [9], nanocrystalline Si films [3], nanostructured gold films [10], and InN nanorods [11]. This model, commonly referred to as the Drude-Smith model, incorporates carrier backscatter and is given by [7]

$$\tilde{\sigma}(\omega) = \frac{\varepsilon_0 \omega_p^2 \tau}{1-i\omega\tau}\left[1+\frac{c}{1-i\omega\tau}\right] \qquad (1)$$

where τ is the carrier scattering time, ω_P is the plasma frequency, and c can vary between $c = 0$ (Drude response with no carrier backscatter) and $c = -1$ (fully localized, 100% carrier backscatter). We find that the Drude-Smith model provides a very useful framework for understanding the nature of terahertz conductivity in nanomaterials.

II. Results

Figure 1 shows the transient THz conductivity measured in photoexcited nanocrystalline silicon films with varying degrees of disorder [3]. In Fig. 1(a), Eq. (1) with $c \approx 0$ fits well to the THz conductivity in an epitaxial silicon-on-sapphire sample. The disorder increases for a poly-nanocrystalline silicon film (Fig. 1(b)), resulting in a suppression of the real part of the THz conductivity at low frequencies due to the onset of some carrier localization. Finally, Fig. 1(c) shows very strong carrier localization with almost complete low-frequency suppression of σ_1 in Si nanocrystal films. It is important to recognize that the terahertz conductivity for the entire sample sequence from free ($c \approx 0$) to localized ($c \approx -1$) behavior fits within the framework of the Drude-Smith model.

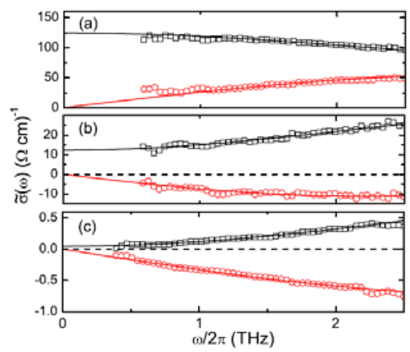

FIG. 1. THz conductivity in photoexcited nanocrystalline Si films [3].

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