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Improved instrumentation for intensity-, wavelength-, temperature-, and magnetic field-resolved photoconductivity spectroscopy



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ABSTRACT

We report instrumentation for photovoltage and photocurrent spectroscopy over a larger continuous range of wavelengths, temperatures, and applied magnetic fields than other instruments described in the literature: $350 \text{ nm} \le \lambda \le 1700 \text{ nm}$, $1.8 \text{ K} \le T \le 300 \text{ K}$, and $B \le 9 \text{ T}$. This instrument uses a modulated monochromated incoherent light source with total power $< 30 \mu$ W in combination with an LED in order to probe selected regions of non-linear responses while maintaining low temperatures and avoiding thermal artifacts. The instrument may also be used to measure a related property, the photomagnetoresistance. We demonstrate the importance of normalizing measured responses for variations in light power and describe a rigorous process for performing these normalizations. We discuss several circuits suited to measuring different types of samples and provide analysis for converting measured values into physically relevant properties. Uniform approaches to measurement of these photoproperties are essential for reliable quantitative comparisons between emerging new materials with energy applications.

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1. Introduction

Photoconductivity is a property of materials that is hugely important to the performance of photodiodes, solar cells, photocatalysts, and other technologies. Photoconductivity measurements can be used to study optical bandgaps [1], carrier lifetimes [2], and other important electronic properties of materials. Measuring photoconductivity as a function of temperature and wavelength is a powerful method for investigating complex band structures [3] and the energies and densities of defects in a material [4]. A related property, the photomagnetoresistance, has been used to study the structure of conduction bands in insulators [5] and spin-dependent recombination [6].

Photoconductivity was discovered serendipitously when a selenium resistor was exposed to ambient light during an electronic measurement [7]. In order to resolve features in photoconductivity spectra, modern photoconductivity experiments often use a monochromatic light source such as a collection of LEDs [8], monochromated lamp [9], or laser [10]. Unlike LEDs or traditional

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http://dx.doi.org/10.1016/j.jssc.2016.02.002 0022-4596/© 2016 Elsevier Inc. All rights reserved. lasers, monochromated lamp sources can produce light over a broad and continuous wavelength range, albeit with relatively less power.

The use of low power, incoherent illumination in photoconductivity measurements is advantageous for several reasons. First, a material's response can vary depending on the coherence of the irradiation and thus an incoherent source is desirable in order to mimic real conditions experienced by devices. Additionally, light energy absorbed by the device under test (sample) and its environment is converted into heat. Large irradiances limit the lowest achievable sample temperature and can complicate the accurate measurement of bulk sample temperature during a measurement by creating temperature gradients within a sample. Further, low light power also helps avoid contaminating thermoelectric effects that stem from temperature gradients and can improve the sensitivity of measurements that involve low-energy transitions by reducing the number of carriers that undergo those transitions by thermal excitation.

Photoresponses of real materials frequently vary with light intensity in a non-linear fashion, especially at very low or very high irradiances, due to the depletion or saturation of defect levels involved in transitions and other effects. One approach to isolating the contribution of one transition to the net photoconductivity is the simultaneous use of multiple light sources. Grimmeiss et al. first demonstrated the use of two light-sources in a constant photoconductivity measurement (CPM) as a means to measure photoionization cross-sections in materials with multiple defect levels [11]. Bube et al. later refined the CPM method and identified several limitations, most importantly that the response of the sample must have an intensity-dependence that is constant for all wavelengths of irradiation [12].

In this work we describe a method in which a frequencychopped low-intensity light source is used simultaneously with a more intense continuous light source in order to probe different regions of a non-linear photoresponse. This method offers several advantages over earlier techniques: (1) the light intensity and photocurrent can be set independently at each wavelength; (2) because only photocurrents and photovoltages resulting from the chopped light source are recorded, thermal effects resulting from the more intense light source may be neglected; (3) the photoconductivity measured at a given intensity is closer to the instantaneous photoconductivity (the change in the conductivity for an infinitesimal change in light intensity) than when more intense sources are used.

We also present the technical details and theory of operation of an instrument for performing these measurements using a chopped light source of wavelength 350 nm $\leq \lambda \leq$ 1700 nm and total incident light power $P < 30 \mu$ W at temperatures 1.8 K $\leq T \leq$ 300 K and magnetic fields $B \leq$ 9 T.

2. Theory of operation

For a given wavelength of light λ , the intensity of light arriving at a certain depth within a sample, $I(x, \lambda)$, is given by the Beer–Lambert law,

$$I(x, \lambda) = I_0(1 - R(\lambda))e^{-\alpha(\lambda)x}$$
(1)

where I_0 is the light intensity arriving at the surface of the sample, $R(\lambda)$ is the reflectivity of the material at that wavelength, α is the absorption coefficient of the material, and x is the depth within the sample. The illumination of a sample may generate free carriers, either directly or through the formation of bound excitons which then dissociate to form free carriers. For an ideal sample under illumination with wavelength λ and thickness t, in the limit of low-intensity, the rate of carrier generation $f(\lambda)$ depends linearly on the incident light power according to:

$$f(\lambda) \propto \int_0^t I(x, \lambda) dx = I_0$$
⁽²⁾

Under constant illumination, the sample will eventually reach a steady state where the rate of optical generation of carriers is equal to the rate of recombination. That is to say, the change in carrier concentration upon illumination will be a function of the rate of free carrier generation and the free carrier lifetime. If one makes the simplifying assumption that the changes in free electron concentration, Δn , and free hole concentration, Δp , are homogenous throughout the illuminated region [13], then the change in carrier concentration can be expressed with

$$\Delta n = f(\lambda)\tau_n \tag{3}$$

where τ_n is the lifetime for free electrons and

$$\Delta p = f(\lambda)\tau_p \tag{4}$$

where τ_p is the lifetime of free holes.

Illuminating a sample will affect the conductivity of the sample according to



Fig. 1. (a) The dashed line indicates the conductivity σ as a function of incident light intensity I_0 for an ideal material. The solid line indicates the same function for a typical photoconductor. (b) One intense and constant light source (I_{DC}) and a smaller, modulated light source (I_{AC}) are used to select a specific region of the photoconductive response.

$$\Delta \sigma = f(\lambda) e(\mu_n \tau_n + \mu_p \tau_p) \tag{5}$$

where $\Delta \sigma$ is the change in conductivity upon illumination (the photoconductivity), *e* is the electron charge, and μ_n and μ_p are the free electron mobility and the free hole mobility, respectively.

In the limit of very small changes in $f(\lambda)$, μ_n , μ_p , τ_n , and τ_p are approximately constant, so it is possible to determine relative changes in $f(\lambda)$ by measuring changes in $\Delta \sigma$ while scanning λ . Abrupt changes in $f(\lambda)$ while

scanning λ indicate band edges or the energies of transitions to and from in-gap defect states. For a material in which $f(\lambda)$ is wellknown, measurements of $\Delta \sigma$ can be used to calculate τ_n and τ_p .

The photoconductivity of many materials is non-linear. At low irradiances $\Delta \sigma$ is suppressed because the trapping of carriers by defects reduces their mobility (Fig. 1a). As I_0 increases the majority of traps will eventually become occupied due to the increasing number of free carriers and the photoconductivity may become approximately linear due to the saturation of traps, As I_0 increases further, the rates of direct recombination and multi-particle scattering increase, decreasing τ and μ respectively. These effects

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