Infrared Physics & Technology 67 (2014) 436–440

Contents lists available at ScienceDirect

Infrared Physics & Technology

journal homepage: www.elsevier.com/locate/infrared

A broadband silicon quarter-wave retarder for far-infrared spectroscopic circular dichroism

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HIGHLIGHTS

- We describe the design of a silicon quarter retarder working in the far infrared.
- The device produces far-infrared light with a high degree of circular polarization.
- We demonstrate its performance by photo-induced magnetospectroscopy of GaAs.

ARTICLE INFO

Article history:
Received 28 July 2014
Available online 16 September 2014

Keywords:
$\frac{1}{4}$ wave retarder
Circularly polarized light
Circular dichroism
Far-infrared magnetospectroscopy
Cyclotron resonance
Excitons

ABSTRACT

The high brightness, broad spectral coverage and pulsed characteristics of infrared synchrotron radiation enable time-resolved spectroscopy under throughput-limited optical systems, as can occur with the high-field magnet cryostat systems used to study electron dynamics and cyclotron resonance by far-infrared techniques. A natural extension for magnetospectroscopy is to sense circular dichroism, i.e. the difference in a material's optical response for left and right circularly polarized light. A key component for spectroscopic circular dichroism is an achromatic $\frac{1}{4}$ wave retarder functioning over the spectral range of interest. We report here the development of an in-line retarder using total internal reflection in high-resistivity silicon. We demonstrate its performance by distinguishing electronic excitations of differing handedness for GaAs in a magnetic field. This $\frac{1}{4}$ wave retarder is expected to be useful for far-infrared spectroscopy of circular dichroism in many materials.

1. Introduction

Infrared spectroscopy is a well-developed technique to study electronic and vibrational properties in a variety of material systems of interest in condensed matter physics, chemistry, and biology. The energy levels associated with electronic transitions or vibrational modes manifest as spectroscopic features at photon energies specific to each material, enabling material identification and characterization. The capabilities of infrared spectroscopy can be greatly enhanced by using polarized light to reveal further details of the excitations. For example, linearly polarized infrared light has been commonly employed to distinguish lattice vibrational modes and electronic excitations along different directions in basic anisotropic crystals.

When the excitation involves the orbital motion of electrons in a magnetic field, or their spin, the absorption of light is usually limited to circularly polarized light of a particular handedness. The difference in the resulting optical response between left and right circularly polarized light is referred to as circular dichroism. The effect can occur for various novel quantum states in condensed matter physics and material science. For example, Landau level spectroscopy in monolayer [1–4], bilayer [2–7], and multilayer [8–10] graphene, graphite [11,12], as well as giant Rashba spin-splitting semiconductors [13] can probe the electronic energy levels quantized by a magnetic field. Circularly polarized light can distinguish transitions with opposite handedness, putting strict constraints on the selection rules for the optical transitions. Faraday and Kerr rotations, caused by different propagation velocities for left and right circularly polarized light in a medium, have been reported in graphene [14]. More intriguingly they have been proposed to experimentally verify the topological magnetoelectric effect in 3D topological insulators, a phenomenon of topological

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http://dx.doi.org/10.1016/j.infrared.2014.09.004
1350-4495/

quantization in the unit of the fine structure constant [15]. Availability of left and right circularly polarized light in principle allows the measurement of the off-diagonal optical conductivity, the quantity underlying the Faraday and Kerr rotations. Another new trend of using circularly polarized light is the study of the valley-dependent circular dichroism in two-dimensional semiconductors, recently observed in MoS_2 [16] and WSe_2 [17] by circularly polarized photoluminescence.

Circularly polarized light is commonly produced (or sensed) using the combination of a linear polarizer and a $\frac{1}{4}$ wave retarder. Linear polarizers made from free-standing wire grids or deposited metal strips on a transparent substrate perform well in the far infrared. On the other hand, $\frac{1}{4}$ wave retarders that exploit birefringent waveplates perform well only over a narrow range of frequencies [18], limiting their usefulness to monochromatic sources such as lasers, as reported in Ref. [19], although careful design based on multiple waveplates has been demonstrated to extend the operating frequency range [20]. An approach for making an achromatic retarder follows the principle of a Fresnel rhomb where the phase shift upon total internal reflection depends only on the refractive index and angle of incidence. However, due to the considerable optical thickness of the device, the choice of material is limited to those having a very low absorption coefficient. In this work, we discuss the development of a Fresnel rhomb-type $\frac{1}{4}$ wave retarder using silicon and demonstrate its capabilities by performing photo-induced magneto-spectroscopy on GaAs.

2. Development and characterization of a silicon $\frac{1}{4}$ wave retarder

2.1. Operating principle and design

Materials that are both transparent and isotropic in the spectral range of interest are necessary for a retarder design. The phase shift between the *s* and *p* polarizations from total internal reflections is determined by the refractive index of the retarder material and the angle of incidence. An optimal design therefore involves careful choices of these two parameters. Maintaining a frequency-independent phase shift implies an almost constant refractive index in the frequency range of interest. We also desire a compact optical system that can be readily added to an existing experimental setup while introducing minimal optical loss. Based on these considerations we have chosen high-resistivity silicon (refractive index $n = 3.42$ in the far infrared) for our retarder. A double-Fresnel rhomb design [Fig. 1(a)] using silicon was proposed by two of us for a far-infrared full Mueller Matrix ellipsometry [21,22]. A consequence of the high refractive index is a rather large phase shift for a single internal reflection. To manage this, an internal reflectance angle of 27° was chosen to yield a $5\pi/8$ phase shift. Thus, for a design where the beam experiences four such reflections [Fig. 1(b)], a total phase shift of $5\pi/2$ ($= 2\pi + \pi/2$), i.e. a net of $\frac{1}{4}$ wave occurs, producing circularly polarized light when the incident light is linearly polarized and oriented to yield equal amplitudes of *s* and *p* reflections.

We implemented this rhomb retarder design using the optical assembly shown in Fig. 1(c). A pair of 50 mm aperture off-axis paraboloidal mirrors focus a collimated incident beam through the linear polarizer and onto the retarder, and re-collimates the transmitted beam back onto the original optical path. The polarizer transmission angle is set to +45° or $-45°$ with respect to the horizontal direction to yield equal amplitudes of *s* and *p* polarized reflected amplitudes inside the rhomb. The linear polarizer's rotation is motorized to allow switching between left or right circularly polarized light. We note that the reflections from the aluminum coated off-axis paraboloids have minimum effects on the polarization state in the far infrared. All optical components are assembled on a 100 mm × 250 mm breadboard, which can be conveniently slid into and out from the beam path. The optics are sufficiently large to work with common commercial FTIR spectrometers having 40 mm diameter optics. In the following demonstrations, the polarization of the incident beam onto the entire circular polarizer instrument (i.e. the retarder plus the upstream polarizer) was set to predominantly vertical. Although this is not required to produce circularly polarized light and led to an additional loss of intensity, it ensured that the intensity transmitted by the system for the left and right circularly polarized light was nearly equal.

2.2. Retarder performance

The retarder system was tested in the far infrared for use mostly below a frequency of 200 cm^{-1}. Fig. 2 shows the transmittance of the retarder between 20 and 250 cm^{-1}, measured at room

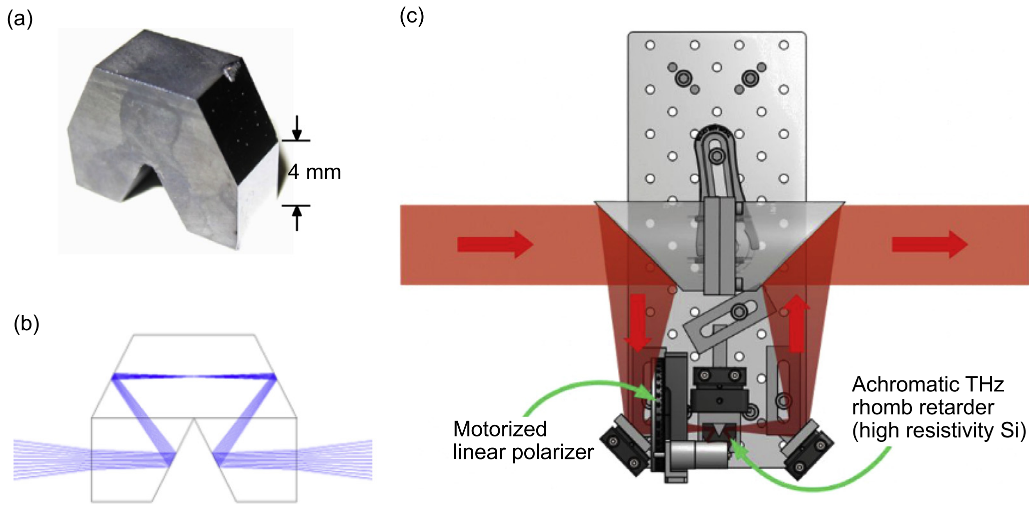

Fig. 1. (a) Double-Fresnel rhomb $\frac{1}{4}$ wave retarder made of high resistivity silicon. The reflecting surfaces in the beam path are mechanically polished to reduce beam scattering. (b) Cross section of the rhomb retarder showing the designed beam path (blue lines). (c) Optical assembly for the circular polarizer. The motorized linear polarizer just upstream of the retarder sets the polarization angle of the incident linearly polarized beam to be ±45° with respect to the horizontal direction (defined according to the right-hand rule), yielding right or left circularly polarized light. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)