ANISOTROPY OF THE OPTICAL TRANSMISSION SPECTRUM IN $BaCuSi_2O_6$

Jaime, M., Crooker S. A., (NHMFL, LANL), Kimura, T. (MST-10, LANL)

Introduction

$BaCuSi_2O_6$ is a quasi two-dimensional spin gap compound comprising weakly-coupled layers of Cu^{2+} dimers with intradimer coupling $J = 4.45$ meV, interdimer coupling $J' = 0.58$ meV and interplane coupling $J'' = 0.116$ meV [1]. A phase boundary separates the low magnetic field quantum paramagnetic state (made up of spin-singlets), from a high field ($23.4 < H < 49$ T) magnetically ordered state, which has been interpreted as a Bose-Einstein condensate of triplet states (triplons) [1].

$BaCuSi_2O_6$ displays a striking optical polarization anisotropy that manifests in our single crystal samples when using a standard optical microscope with bottom illumination (Figure 1). In order to quantify this effect, we measured the optical transmission spectrum of polarized light from ultraviolet to infrared wavelengths, using a polarized white light source, spectrometer, and CCD camera. The results of the experiment are displayed in Figure 2. Regardless of polarization, $BaCuSi_2O_6$ exhibits strong optical absorption (no transmission) in the range 475-600 nm (blue-green through orange wavelengths). However, the transmitted optical spectrum shows dramatic anisotropy in the 600-900 nm (red-IR) range when the polarization of the incident light is rotated from parallel to perpendicular to the Cu^{2+} bilayers. A less dramatic and opposite effect is observed in the 350-475 nm (UV-blue) range. Clearly, intra-bilayer electronic excitations strongly absorb light at red wavelengths (if the light is polarized in the plane of the bilayers), making the sample appear deep blue/violet when viewing transmitted light through a bottom illuminated sample. Light polarized along the inter-bilayer direction, on the other hand, is transmitted in the red, and also in the blue, making the sample appear reddish-purple. Consequently the peculiar purple coloration displayed by $BaCuSi_2O_6$ is the result of both strong absorption in the green region of the spectrum, and anisotropy. The origin of the particular absorption resonances are, at this time, not understood.

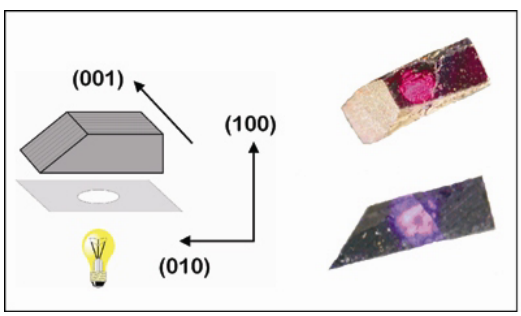

Figure 1: Oriented single crystal sample of $BaCuSi_2O_6$ as observed with an optical microscope with bottom illumination.

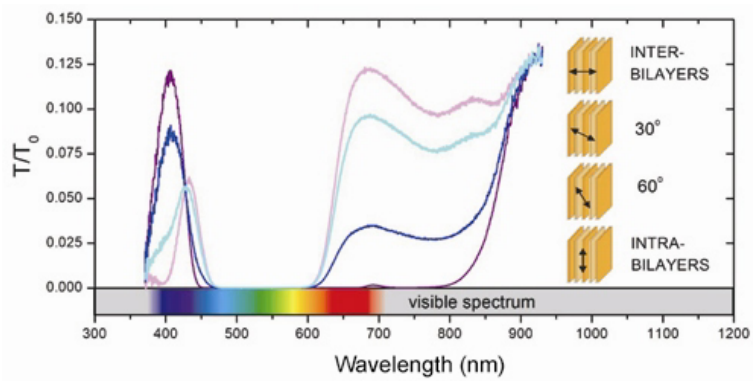

Figure 2: Transmission spectrum measured at four different angles between the electrical field vector in the polarized light beam and the Cu bilayers as indicated.

Acknowledgements

Experiments performed at the National High Magnetic Field Laboratory were supported by the NSF through Co-operative Grant No. DMR-90162411 and MRI Grant No. 0079641, the State of Florida, and the Department of Energy.

References

[1] M. Jaime *et al*., Phys. Rev. Lett. **93**, 0872031 (2004).