Magnetoelectric coupling in 4,4′-stilbenedinitrene

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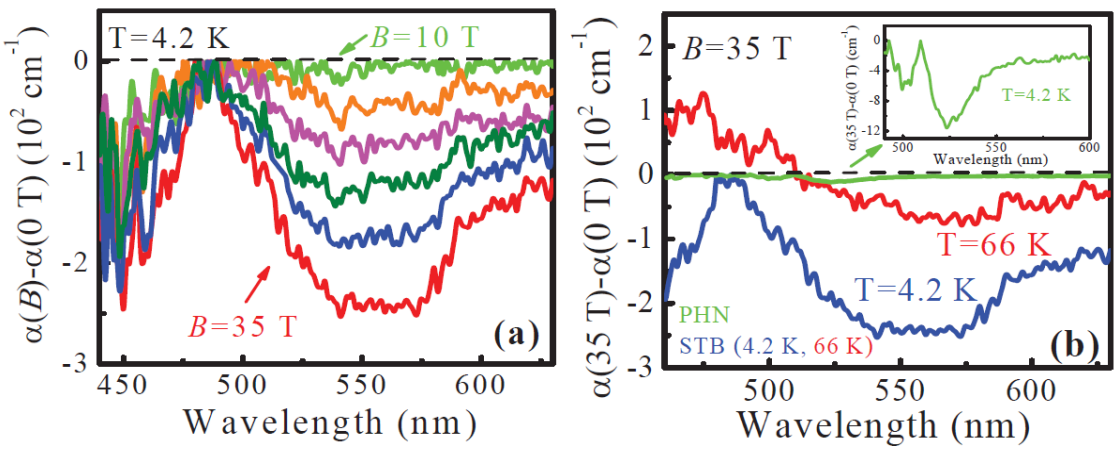
Introduction

Magnetoelectric coupling in materials like multiferroics, dilute magnetic semiconductors, and topological insulators has attracted a great deal of attention, although most work has been done in the static limit. Magneto-optical spectroscopy offers a way to investigate the dynamics of charge-spin coupling, an area where there has been much less effort. We recently sought to extend these techniques beyond complex oxides, specifically to open-shell systems with kinetically trapped spin states such as those that exist in many radicals and biradicals.[1,2] This strategy combines high field techniques with the advantages of low energy scale organic magnets to generate potentially substantial magnetoelectric coupling.



(a) Absorption difference spectra,α(B)-α(0 T), o 4,4′-stilbenedinitrene at 4.2 K as a function of magnetic field: B = 10, 15, 20, 25, 30, 35 T. (b) Absorption difference spectrum of 4,4′-stilbenedinitrene (STB) and 1,4-phenylenedinitrene (PHN)[3] biradicals at full field, α(B = 35 T)-α(0 T), at 4.2 and stilbene biradical at 66 K. Inset: Close up view of the absorption difference spectrum of 1,4-phenylenedinitrene at4.2 K for comparison. Clearly, conjugation length can be used to amplify and tune the magnetochromic response in molecular systems.[4]

Results and Discussion

To test these ideas, we investigated the optical properties of 4,4′-stilbenedinitrene at low temperature and in high magnetic fields and compared the results with complementary first principles calculations. Both physical tuning parameters allow us to manipulate the singlet-triplet equilibrium, and by so doing, control the optical contrast (which is on the order of -2.5×10^2 cm^{-1} at 555 nm and 35 T). Moreover, analysis of the magneto-optical response using a combined population and Beer's law framework reveals the singlet-triplet spin gap and identifies particular features in the absorption difference spectrum as deriving from singlet or triplet state excitations. These findings deepen our understanding of coupling in open shell molecules and show how chemical structure modification can modulate charge-spin interactions in organic biradicals.

Acknowledgements
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References
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