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Form Approved
OMB No. 0704-0188

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1. REPORT DATE (DD-MM-YYYY) 08-07-2011		2. REPORT TYPE Final	3. DATES COVERED (From - To) August 15 2009 - February 14 2011		
4. TITLE AND SUBTITLE Exploring Electrical and Magnetic Resonances from Coherently Correlated Long-Lived Radical Pairs towards Development of Negative Refractive-Index Materials			5a. CONTRACT NUMBER		
			5b. GRANT NUMBER FA9550-09-1-0624		
			5c. PROGRAM ELEMENT NUMBER		
6. AUTHOR(S) Bin Hu			5d. PROJECT NUMBER		
			5e. TASK NUMBER		
			5f. WORK UNIT NUMBER		
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Department of Materials Science and Engineering, University of Tennessee, Knoxville, TN 37996			8. PERFORMING ORGANIZATION REPORT NUMBER		
9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) Air Force Office of Scientific Research 875 North Randolph Street Suite 325, Room 3112 Arlington, VA 22203			10. SPONSOR/MONITOR'S ACRONYM(S)		
			11. SPONSOR/MONITOR'S REPORT NUMBER(S) AFRL-OSR-VA-TR-2012-0734		
12. DISTRIBUTION / AVAILABILITY STATEMENT distribution A					
13. SUPPLEMENTARY NOTES					
14. ABSTRACT During the project period from August 15, 2009 to February 14, 2011, the major achievements include (1) generation of radical pairs by using photoexcitation, (2) determination of magnetic and electric signatures of radical pairs, (3) modification on permittivity and permeability by using radical pairs, (4) mutual coupling between permittivity and permeability, and (5) high Seebeck effect from hybrid metal/polymer/metal thin-film device. In summary, this project has established two new experimental approaches: (i) using radical pairs for the development of molecular metamaterials and (ii) using hybrid metal/polymer/metal thin-film design for the development of highly efficient thermoelectric devices.					
15. SUBJECT TERMS					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			19b. TELEPHONE NUMBER (include area code)

Project title: Exploring Electric and Magnetic Resonances from Radical Pairs towards Development of Negative Refractive Index Molecular Materials

Grant number: FA9550-01-0624

Project period: August 15, 2009 – February 14, 2011

Principle investigator: Bin Hu – University of Tennessee

Research collaborator: Augustine Urbas – Air Force Laboratory – WPAFB

I. Abstract:

This project has developed two new approaches: (i) using radical pairs to modify permittivity and permeability towards the development of molecular metamaterials and (ii) using hybrid metal/polymer/metal thin-film design to separately control electrical and thermal conduction for the development of highly efficient thermoelectric devices. The major achievements are reflected by the following successful experimental discoveries.

1. Radical pairs have been generated with tunable electric and magnetic dipole moments in donor-acceptor molecular systems under photoexcitation.
2. Magnetic and electric signatures of radical pairs have been determined by photoinduced EPR (electron paramagnetic resonance) and photoinduced dielectric measurement.
3. Both permittivity and permeability have been modified by using radical pairs in molecular systems. The permittivity can be changed dramatically after the radical pairs are generated under photoexcitation. The resonant frequency of photoinduced EPR can be shifted after the radical pairs are generated under photoexcitation.
4. The magnetocapacitance measurements have discovered that permittivity and permeability can be coupled by photogenerated radical pairs. This coupling provides a foundation for realizing electromagnetic resonance towards the development of molecular metamaterials.
5. High Seebeck effects have been achieved by using hybrid metal/polymer/metal thin-film design.

II. Selected major discoveries:

1. Magneto-optical response has been observed from radical pairs by magnetic field effects of photoluminescence in donor-acceptor (TPD+BBOT) system (Fig. 1)^[1].

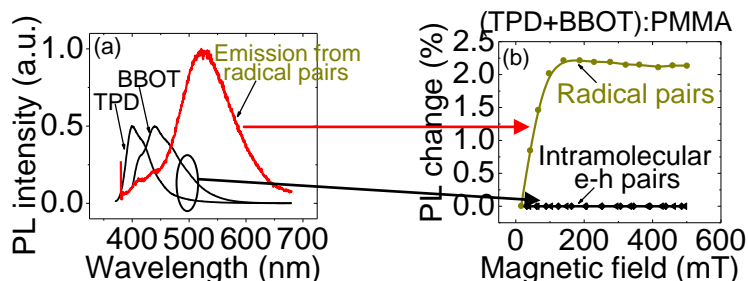


Fig. 1 Photoluminescence spectrum from radical pairs (a) and photoluminescence intensity as a function of magnetic field for radical pairs (b). Note: photoluminescence from non radical pairs are in-sensitive to magnetic field.

This experimental study has found that the photoluminescence from radical pairs is sensitive to magnetic field. This experimental phenomenon forms a foundation to use radical pairs for modification of permittivity and permeability.

2. Magnetic signature of radical pairs has been determined by photoinduced EPR in donor-acceptor Ir65+TCNB system (Fig. 2)^[2].

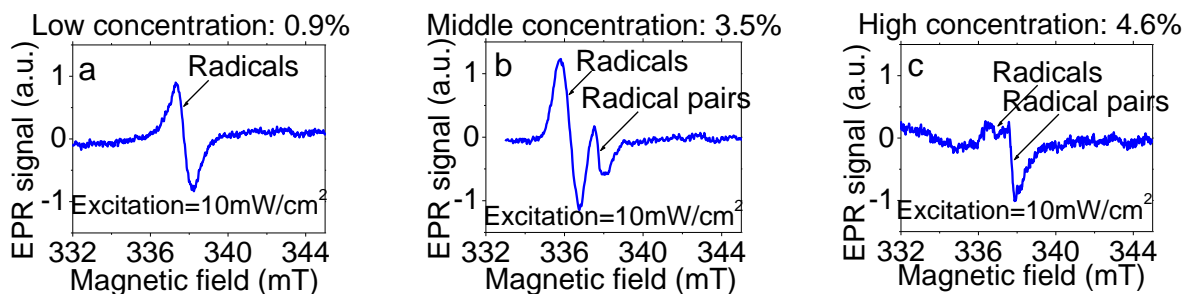


Fig. 2 Photoinduced EPR observed from donor-acceptor (Ir65+TCNB) with low (a), middle (b), and high (c) solution concentrations under photoexcitation of 10 mW/cm².

The photoinduced EPR results show the following remarks.

- (1) Low-concentration liquid shows only one peak, suggesting the existence of radicals.
- (2) Middle-concentration liquid shows two well-resolved peaks, suggesting the existence of both radicals and radical pairs.
- (3) High-concentration liquid shows two un-resolved peaks, suggesting the existence of high-density radicals and radical pairs.

- We have successfully measured dynamic processes of radical pairs by using transient photoinduced absorption at WPAFB (Dr. Urbas group) by using light-emitting radical pairs formed in donor-acceptor (Pyrene+DMA) system^[3].

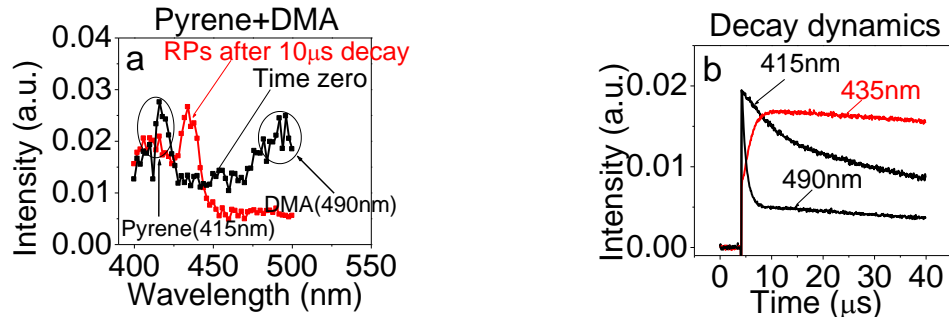


Fig. 3 **a**: Photoinduced absorption spectra for excitons in Pyrene (415 nm), excitons in DMA (490 nm), and radical pairs (RPs) (435 nm) at different time scales. **b**: Dynamic behavior of radical pairs (435 nm) as compared to the excitons in Pyrene (415 nm) and the excitons in DMA (490 nm).

We can see in time-dependent measurement that the density of radical pairs (435 nm) increases as the excitons decrease in density in Pyrene (415 nm) and DMA (490 nm). This successful measurement provides important information to determine the electromagnetic response time of radical pairs for the development of permittivity and permeability resonances.

- We discovered that photogenerated radical pairs can generate a coupling between permittivity and permeability (Fig. 4)^[4].

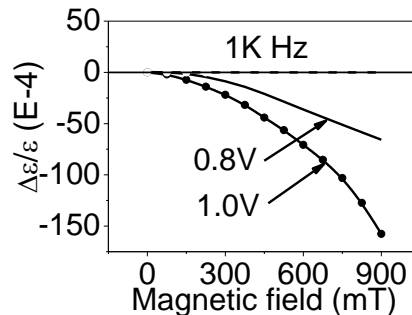


Fig. 4 Permittivity changes as a function of magnetic field in radical-pair system.

This experimental result indicates that radical pairs are promising approach to realize resonance in both permittivity and permeability.

- We discovered a new approach to develop high Seebeck effects by using common metal and conducting polymer materials based on hybrid metal/polymer/metal thin-film design through interface control (Fig. 5)^[5]. The obtained Seebeck effects are comparable to highly efficient inorganic semiconductors.

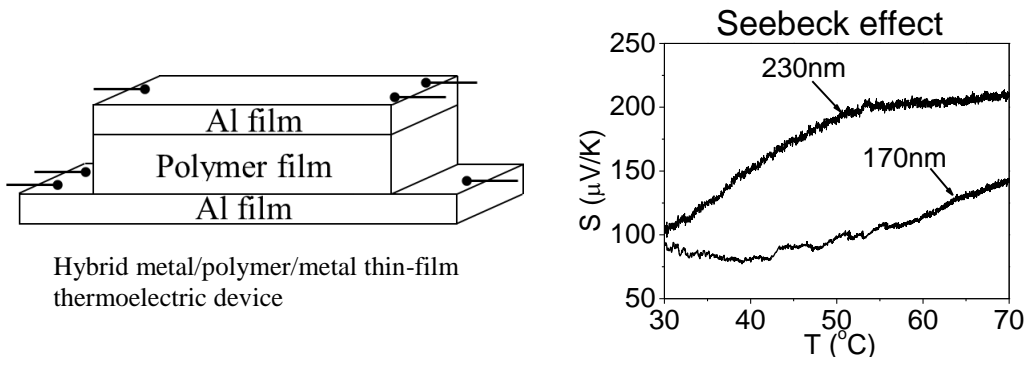


Fig. 5 Hybrid metal/polymer/metal thin-film thermoelectric device and Seebeck effect.

In summary, this project has established two new approaches: (i) using radical pairs to modify permittivity and permeability for the development of molecular metamaterials and (ii) using hybrid metal/polymer/metal thin-film design to separately control electrical and thermal conductions for the development of highly efficient thermoelectric devices.

III. Archival publications

- [1] Magnetic Field Effects in Organic Semiconductors
Bin Hu*, Liang Yan, and Ming Shao,
Adv. Mater. **21**, 1500, 2009.
- [2] Changing Inter-molecular Spin-Orbital Coupling for Generating Magnetic Field Effects in Phosphorescent Organic Semiconductors
Liang Yan, Ming Shao, Carlos F. O. Graeff, Ivo Hummelgen, Bin Hu*
Physical Review Letters, Submitted.
- [3] Dynamic behavior of photogenerated radical pairs for application of molecular metamaterials
Liang Yan, Jon Slagle, Augustine Urbas, and Bin Hu*, to be published.
- [4] Room-Temperature Multiferroic Phenomenon in Organic Semiconductors
Huidong Zang, Liang Yan and Bin Hu*, to be published.
- [5] High Seebeck Effects from Hybrid Metal/Polymer/Metal Thin-Film Devices”
Liang Yan, Ming Shao, Hsin Wang, Doug Dudis, and Augustine Urbas, and Bin Hu*
Adv. Mater. In press