ELSEVIER

14 July 1995

Chemical Physics Letters 241 (1995) 89–96

CHEMICAL
PHYSICS
LETTERS

Measurement of absolute photoluminescence quantum efficiencies in conjugated polymers

N.C. Greenham [a], I.D.W. Samuel [a], G.R. Hayes [a], R.T. Phillips [a], Y.A.R.R. Kessener [b], S.C. Moratti [c], A.B. Holmes [c], R.H. Friend [a]

[a] *Cavendish Laboratory, Madingley Road, Cambridge CB3 0HE, UK*
[b] *Philips Research Laboratories, Prof. Holstlaan 4, 5656 AA Eindhoven, The Netherlands*
[c] *Melville Laboratory for Polymer Synthesis, Pembroke Street, Cambridge CB2 3RA, UK*

Received 31 March 1995; in final form 15 May 1995

Abstract

Measurements of absolute photoluminescence (PL) efficiencies have been performed for solid films of several conjugated polymers commonly used for electroluminescence. In poly(*p*-phenylenevinylene) (PPV), a PL efficiency of 0.27 is measured in samples which show an initial PL decay time-constant of 320 ps. These values indicate that photoexcitation in PPV produces intra-chain singlet excitons with a high quantum yield. The PL efficiencies of derivatives of PPV have been investigated, and efficiencies in excess of 0.4 have been measured for cyano-substituted PPVs.

1. Introduction

The luminescence properties of conjugated polymers are of considerable interest, both because of the fundamental information that can be obtained about exciton formation and decay, and because of the potential applications for conjugated polymers as the emissive material in light-emitting diodes (LEDs) [1,2]. Luminescence in conjugated polymers is believed to be the result of radiative decay of singlet excitons. Competing non-radiative processes provide additional means of decay, and therefore reduce the efficiency of luminescence. Possible non-radiative mechanisms in the solid state include inter-chain processes (e.g. excimer formation), and quenching of excitons by extrinsic or conformational defects. If radiative and non-radiative decay are monomolecular processes with rates τ_r^{-1} and τ_{nr}^{-1}, respectively, the overall luminescence decay will be exponential, with a lifetime, τ, given by

$$\tau^{-1} = \tau_r^{-1} + \tau_{nr}^{-1}. \qquad (1)$$

The efficiency for radiative decay of singlet excitons is then given by

$$q = \tau/\tau_r. \qquad (2)$$

The efficiency of radiative decay of singlet excitons, q, sets an upper limit on the quantum efficiency which can be obtained in a polymer LED.

In a photoexcitation experiment, a useful figure which can be measured is the photoluminescence (PL) efficiency, defined as the number of photons emitted in photoluminescence per absorbed photon.

SSDI 0009-2614(95)00584-6

This figure depends both on q and on the fraction, η, of absorbed photons which lead to the formation of singlet excitons. The PL efficiency, Q, is given by

$$Q = \eta q. \tag{3}$$

On the basis of picosecond stimulated-emission experiments, Rothberg and co-workers have recently suggested that in poly(p-phenylenevinylene) (PPV), only about 10% of the photoexcited species are singlet excitons, and that the rest are spatially indirect polaron pairs which do not result in radiative decay [3]. In this case, the measured PL efficiency would be only 10% of the efficiency for radiative decay of singlet excitons, corresponding to a value of $\eta = 0.1$. In this Letter, we present direct measurements of PL efficiency for a number of conjugated polymers, and use these results to address the issues of exciton formation and decay in this class of materials.

In contrast to measurements of PL efficiencies in solution, measurements on thin solid films are not straightforward, since the angular distribution of the emitted light is highly sensitive to the refractive index of the material, and to the orientation of emitting dipoles within the film [4,5]. Estimates of absolute PL efficiencies from a single measurement of light output in the forward direction [6] must therefore be treated with considerable caution.

Information about the efficiency of radiative decay of singlet excitons can be obtained from measurements of the overall PL lifetime, τ. In order to extract a value of q from these measurements, however, it is necessary to obtain an independent estimate of the natural radiative lifetime, τ_r. Such estimates have been made by determining the radiative lifetime of the polymer or a model oligomer in solution [7,8] from the measured quantum efficiency and total lifetime in solution. Given certain assumptions, the radiative lifetime can be related to the absorption and emission spectra using the Strickler–Berg relationship [9],

$$\tau_r^{-1} = \frac{8\pi n_{\mathrm{ref}}^2}{c^2} \langle \nu^{-3} \rangle^{-1} \int \frac{\sigma(\nu)\,\mathrm{d}\nu}{\nu}, \tag{4}$$

where

$$\langle \nu^{-3} \rangle^{-1} = \frac{\int L(\nu)\,\mathrm{d}\nu}{\int \nu^{-3} L(\nu)\,\mathrm{d}\nu}. \tag{5}$$

n_{ref} is the refractive index, $\sigma(\nu)$ is the absorption cross section per absorbing species at frequency ν, and $L(\nu)$ is the emission spectrum as a function of photon frequency, ν. Assuming that the same intrachain transition is responsible for emission in solution and in the solid state, the radiative lifetime in the solid state can be estimated from the solution radiative lifetime using the measured refractive indices and emission spectra. In practice, the PL decay is frequently non-exponential [7,8,10–12], and refractive indices are difficult to determine accurately. Changes in chain conformation and the effects of inter-chain interactions can also affect the character of the radiative transition. Due to these difficulties, estimates of q from lifetime measurements can only be regarded as rough estimates.

In this Letter, measurements of the absolute PL efficiency in solid films of conjugated polymers are performed using an integrating sphere to collect the emitted light. Until now, this technique has only been applied to a limited number of conjugated polymers [13,14]. The intention here is to study a wide range of materials commonly used for EL devices, to describe the measurement technique in detail, and to discuss the implications for the photophysics of conjugated polymers. We compare the results for PPV with PL lifetime measurements, and estimate the value of the branching ratio, η.

2. Experimental details

An integrating sphere is a hollow sphere, coated on the inside with a diffusely reflecting material. The flux received at an aperture in the sphere (the exit port) is proportional to the total amount of light produced within the sphere, irrespective of its angular distribution [15]. A number of experimental precautions are necessary to obtain reliable PL efficiencies from integrating sphere measurements, as described below.

A custom-built, 10 cm diameter integrating sphere manufactured by Labsphere was used. Thin films of conjugated polymer were formed by spin-coating onto Spectrosil discs of thickness 1–2 mm, and diameter 13–20 mm. Optical densities at the excitation wavelength were at least 1.0, and preferably

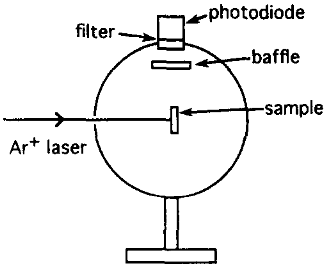


Fig. 1. Experimental arrangement for integrating sphere measurements.

greater than 2.0. These samples were mounted at the centre of the integrating sphere, as shown in Fig. 1. Light from an argon-ion laser was incident onto the sample through a small hole in the sphere wall. Typical laser powers were in the range 0.1–0.5 mW, with a beam diameter at the sample of approximately 1 mm. Conditions of near-normal incidence were used, but the beam reflected from the sample surface was not allowed to escape directly though the entrance hole. The luminescence was measured using a calibrated silicon photodiode (Graseby-UDT 221) at the exit port. A diffusely reflecting baffle was positioned between the sample and the exit port in order to prevent luminescence from reaching the detector directly. A suitable filter was placed in front of the detector to absorb the excitation laser wavelength, whilst transmitting the luminescence. When using Schott glass filters, it was found that fluorescence from the filter itself contributed a significant fraction of the detected signal. This problem was alleviated by using Kodak Wratten gelatin filters, which were found to be much less fluorescent.

The incident laser power was measured with sample and filter removed. To calculate a PL efficiency, it is important to know how much of the incident light is absorbed within the polymer. The normal reflectance and transmittance of each sample were measured at the excitation wavelength. Laser light scattered diffusely by the sample was neglected, but this was not thought to introduce a significant error. Some of the laser light which is not absorbed at the first attempt is absorbed by the sample after reflection from the walls of the integrating sphere. The amount of luminescence due to this subsequent absorption was determined by measuring the luminescence detected with the sample in the integrating sphere but with the laser incident on the wall of the sphere rather than on the sample. The quantum efficiency, neglecting the spectral variation of the system response, was given by

$$x = \frac{X_{\text{sample}} - (R+T)X_{\text{sphere}}}{(1-R-T)X_{\text{laser}}}, \tag{6}$$

where R is the reflectance, T is the transmittance, and X_{laser} is the signal measured with the laser incident on the sphere with no sample or filter. X_{sample} and X_{sphere} are the signals measured with the sample and filter in place, with the laser incident on the sample and the sphere wall, respectively.

The spectral response of the sphere was determined using a tungsten lamp and a parallel-detection CCD spectrograph (Oriel Instaspec IV) with an optical fibre input. The spectrum of the lamp, $S_{lamp}(\lambda)$, was first measured outside the sphere. The lamp was then used to illuminate the sphere through the laser entrance hole, and the spectrum, $S_{sphere}(\lambda)$, was measured at the exit port. The correction factor for the spectral response is given by

$$y = \int \frac{S_{\text{sphere}}(\lambda)L(\lambda)G(\lambda)F(\lambda)}{S_{\text{lamp}}(\lambda)}\,\mathrm{d}\lambda \times \left(\frac{S_{\text{sphere}}(\lambda_{\text{ex}})G(\lambda_{\text{ex}})}{S_{\text{lamp}}(\lambda_{\text{ex}})}\int L(\lambda)\,\mathrm{d}\lambda\right)^{-1}, \tag{7}$$

where $L(\lambda)$ is the emission spectrum, $G(\lambda)$ is the quantum efficiency of the photodiode, λ_{ex} is the excitation wavelength, and $F(\lambda)$ is the transmission of the filter. The PL efficiency is then simply given by x/y.

For PPV, the decay of the PL was studied on a picosecond timescale by up-conversion in a nonlinear crystal. The PL efficiency of the sample was measured before and after the time-resolved measurement to exclude the possibility of sample degradation. For the time-resolved measurement, excitation was provided at 405 nm using the frequency-doubled output of a mode-locked Ti–sapphire laser, giving 100 fs pulses at a repetition rate of 76 MHz. The average power was approximately 2 mW, with a beam diameter of 0.4 mm, and the overall time-resolution was approximately 200 fs.

3. Results and discussion

The polymers studied here were PPV, poly(2-methoxy-5-(2′-ethyl-hexyloxy)-*p*-phenylenevinylene) (MEH-PPV), poly(3-hexylthiophene) (P3HT), and two cyano-substituted dialkoxy-PPVs (CN-PPV and MEH-CN-PPV), as shown in Table 1. Two different samples of poly(hexylthiophene) were used denoted P3HT-a and P3HT-b, containing 80% and 100% head-to-tail linkages respectively. MEH-PPV, P3HT and CN-PPV films were formed by spin coating from chloroform; MEH-CN-PPV was spin-coated

Table 1
Chemical structure and synthesis of the polymers investigated

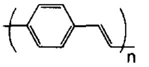
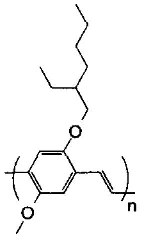
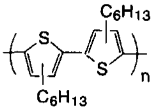
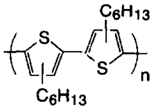
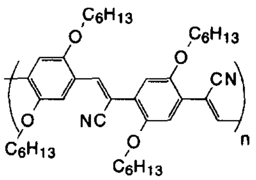
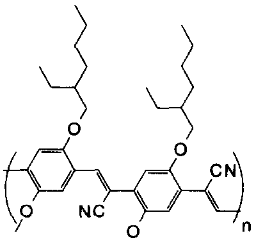
Polymer	Structure	Synthesis
PPV		[16]
MEH-PPV		[17]
P3HT-a (80% head-to-tail)		[18]
P3HT-b (regioregular)		[19,20]
CN-PPV		[21]
MEH-CN-PPV		[22]

Table 2
PL efficiencies, errors and excitation wavelengths for the polymers shown in Table 1

Polymer	PL efficiency	Error	Excitation
PPV	0.27	±0.02	458 nm
MEH-PPV	0.10–0.15	±0.01	488 nm
P3HT-a	0.018	±0.004	488 nm
P3HT-b	0.020	±0.004	488 nm
CN-PPV	0.35–0.46	±0.02	488 nm
MEH-CN-PPV	0.48	±0.02	488 nm

from toluene solution. PPV was formed by the THT precursor route; the precursor was heated at 280°C for 12 h under dynamic vacuum at a pressure of less than 5×10^{-6} mbar. The results obtained for these polymers are shown in Table 2, along with the excitation wavelength used. It is difficult to assess the scale of any systematic errors present in this experiment; the errors shown represent a rough estimate of the likely error due to the measurement technique. The values obtained on different films made from the same batch of polymer were consistent, provided that the films were kept in the dark and under nitrogen until measurement. For MEH-PPV and CN-PPV where several batches were available, a range of efficiencies was measured, as shown in Table 2. The PL efficiency in PPV is known to depend strongly on the conditions used for conversion of the precursor to the final polymer [10,23,24]. We have not yet systematically studied the effect of these conditions on the absolute PL efficiency; the conditions used here are chosen to produce well-converted samples.

A significant decrease in efficiency occurred if the films were stored under ambient laboratory conditions. In MEH-PPV, for example, the PL efficiency was reduced from 0.15 to 0.03 over a period of 1 week, and, during measurement in air, a noticeable decay of the luminescence was observed over a period of tens of seconds. The values shown in Table 2 were measured after less than 5 s of excitation. The exact rate of decay depended strongly on the polymer, the laser power, and whether the excitation was through the glass or directly onto the polymer. The decay could be significantly reduced by flushing the sphere continuously with nitrogen. Under these conditions, the PL efficiency of a PPV sample, for example, decayed by 8% of its initial value after excitation at 0.5 mW directly onto the polymer for 5 min. The decay during measurement, and under storage, is consistent with quenching of luminescence due to defects formed by photo-oxidation [12].

The PL efficiencies for the PPV-based polymers measured here are encouragingly high for polymers in the solid state. The value of 0.27 for PPV, in particular, is higher than the value measured in MEH-PPV, and is significantly larger than the value estimated by Lemmer et al. for PPV [11]. The PL efficiency of MEH-PPV in solution has been estimated to be between 0.20 and 0.35 [7,25]. (Reported values of 0.85 [8] have since been revised [25].) The values of up to 0.15 measured here for solid films suggest that extra non-radiative channels are available in the solid state. The values measured here are similar to the PL efficiencies of 11%–13% reported for solid films of poly(2-methoxy-5-decyloxy-*p*-phenylenevinylene) [13]. The PL efficiencies in excess of 0.40 for the cyano-substituted materials are consistent with the high electroluminescence (EL) efficiencies obtained in these materials [21,22]. In P3HT-a the PL efficiency is much lower than in the PPV-based polymers, consistent with the low EL efficiencies obtained in devices made from this material [26]. The photophysical properties of the poly(alkylthiophene)s are sensitive to the regioregularity of the polymer. Xu and Holdcroft have found that increasing the proportion of head-to-tail linkages in P3HT from 50% to 80% causes a factor of 4 decrease in PL efficiency due to better inter-chain ordering leading to an enhancement of the formation of non-emissive excimers [27]. The P3HT-a used here was synthesised by oxidative coupling of hexylthiophene units, and contains approximately 80% head-to-tail linkages [18]. The regioregular material, P3HT-b, was synthesised using a Grignard coupling reaction [19], and contains almost 100% head-to-tail linkages [20]. The results shown in Table 2 indicate that increasing the fraction of head-to-tail linkages from 80% to 100% causes no significant difference to the PL efficiency.

As discussed earlier, the PL efficiency is given by the product of the branching ratio, η, with the efficiency of radiative decay of the singlet exciton, q. Since q cannot be more than unity, the value of 0.27 obtained for the PL efficiency in PPV gives a lower limit on the branching ratio, η. In order to obtain a better estimate of η, it is useful to estimate q by studying the decay of the PL on a picosecond timescale. In view of the wide range of PL decay rates reported for PPV [7,10–12,23,28], it is necessary to use the same samples for PL efficiency and PL decay measurements. Using the technique described in Section 2, the PL in PPV at 2.25 eV was followed from 0 to 500 ps, and good fits were obtained with a mono-exponential decay with a time-constant of 320 ps.

PL lifetimes of 1.2 ns have been measured by Yan et al. for PPV samples with a low density of oxygen defects, implying a radiative lifetime of at least 1.2 ns [12]. Taking $\tau_r = 1.2$ ns gives a value of $q = 0.28$. The measured PL efficiency of 0.27 therefore suggests a value of η close to unity, implying that the species produced by photoexcitation is predominantly the singlet exciton. We note, however, that the radiative lifetime is sensitive to the size of the emitting chromophore, and may therefore depend on the degree of conjugation of the material studied. If the radiative lifetime in the material studied here is significantly less than in the material studied by Yan et al., then a smaller value of η would be required. An additional source of possible error here is the difference in excitation wavelength between the PL efficiency measurement and the time-resolved measurement. Changing the excitation wavelength from 458 to 405 nm has, however, been found to have only a very small effect on the PL efficiency in high-quality PPV [29,30]. We note that the value of η estimated here is significantly larger than the value of $\eta = 0.1$ estimated by Yan et al. on the basis of picosecond stimulated-emission measurements [3].

The absolute values of PL efficiency reported here provide interesting comparisons between materials. It has generally been taken to be the case that increasing the volume fraction of inactive side-chains (alkyl, alkoxy, etc.) gives an increase in PL efficiency by reducing the rate of diffusion of excitons to quenching sites. Our results, however, show PPV to be more efficient than MEH-PPV. Gettinger et al. [25] have reported that in solution increasing the chain stiffness increases PL efficiency, with MEH-PPV less efficient than polymers with bulkier cholestanoxy side-groups. We propose that conformational defects in the polymer chain may facilitate non-radiative decay of excitons, both in solution and in the solid state. This would account for the relatively low PL efficiencies in solution for MEH-PPV in comparison with more rigid polymers. In the solid state, polymers with low crystallinities and with low glass transition temperatures, such as MEH-PPV, are likely to contain a higher density of conformational defects than PPV, which is relatively crystalline [31].

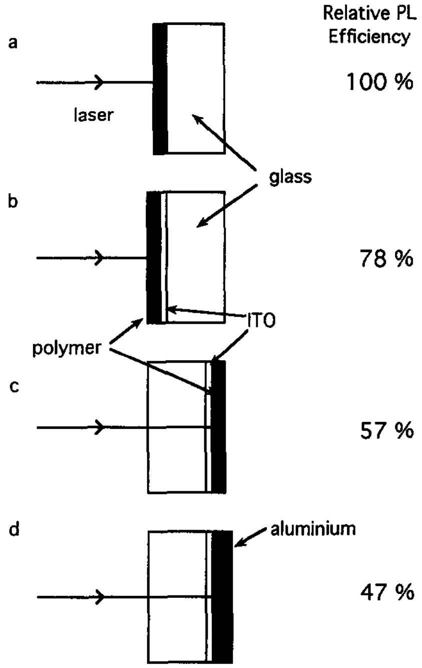


Fig. 2. Relative PL efficiencies for CN-PPV in various structures. (a) polymer on glass substrate; (b) polymer on ITO, excited directly onto polymer; (c) polymer on ITO, excited through glass substrate; (d) polymer on ITO, covered with aluminium. Efficiencies are shown as a percentage of the value measured for case (a). Typical values of the absolute PL efficiency in case (a) are shown in Table 1. Excitation wavelength = 488 nm.

When comparing PL and EL efficiencies, it is important to take into account the effect of the local environment on the PL efficiency of a molecular emitter. Drexhage [4] and others have shown that the presence of dielectric or metal interfaces can significantly alter the radiative lifetime. In general, the effect will depend on the distances of the emitting dipoles from the various interfaces, and on their orientation. For a dipole oscillating close to, and parallel to a metal interface, for example, the radiative lifetime will be increased, leading to a reduced

PL efficiency. These effects occur at distances comparable to the wavelength of the emitted light, and are therefore likely to be important in typical LED structures. In addition, within about 10 nm of an interface, direct quenching of excitons can occur, leading to an increased non-radiative decay rate [32].

As an illustration, PL efficiency measurements were performed using CN-PPV films (thickness 200 nm) which were prepared on glass substrates coated with indium–tin oxide (ITO), as shown in Fig. 2. ITO is commonly used as an electrode material in polymer LEDs. Measurements were also made on a film of CN-PPV sandwiched between ITO and aluminium layers, forming a structure similar to a simple LED. The PL efficiencies measured in various configurations are shown in Fig. 2, and are lower than the values measured for an identical polymer film on an un-coated glass substrate. In cases (c) and (d), the luminescence is generated close enough to one of the interfaces for exciton quenching to be important, in addition to any changes in radiative lifetime. In case (b), however, the amount of light generated within 10 nm of the polymer/ITO interface is very small. An increase in radiative lifetime due to optical interference of the emitted light must therefore be responsible for the decrease in PL efficiency in this case. Although case (d) resembles an LED structure, it is not possible to draw direct comparisons with LED efficiencies since the spatial distribution of luminescence is not the same here as in an LED. These results, however, illustrate the importance of understanding and controlling the location of exciton formation in polymer LEDs.

4. Conclusions

The measurement of PL efficiencies in solid films of conjugated polymers has been described, paying particular attention to the effect of laser light transmitted or reflected by the sample. A number of conjugated polymers commonly used for electroluminescence have been investigated, and PL efficiencies in excess of 0.4 have been measured for cyano-substituted PPV derivatives. The effect of dielectric interfaces on PL efficiencies has also been demonstrated. The measured PL efficiency in PPV sets a lower limit of 0.27 on η, the fraction of absorbed photons which produce singlet excitons, and the measured PL lifetime of 320 ps suggests that η is in fact much larger than 0.27. Taking a radiative lifetime of 1.2 ns gives a value of η close to unity, suggesting that intra-chain singlet excitons are the main product of photoexcitation in PPV.

Acknowledgements

We would like to thank R.W. Jackson, K.A. Murray, J. Rühe, G. Wegner, and Cambridge Display Technology for provision of materials studied in this Letter. This work was partly supported by the Commission of the European Union ESPRIT project LEDFOS-8013 and by the Engineering and Physical Sciences Research Council, UK. We are also grateful to Clare College, Cambridge (NCG) and Christ's College, Cambridge (IDWS) for support.

References

[1] J.H. Burroughes, D.D.C. Bradley, A.R. Brown, R.N. Marks, K. Mackay, R.H. Friend, P.L. Burn and A.B. Holmes, Nature 347 (1990) 539.

[2] D. Braun and A.J. Heeger, Appl. Phys. Letters 58 (1991) 1982.

[3] M. Yan, L.J. Rothberg, F. Papadimitrakopoulos, M.E. Galvin and T.M. Miller, Phys. Rev. Letters 72 (1993) 1104.

[4] K.H. Drexhage, in: Progress in optics, Vol. 12, ed. E. Wolf (North-Holland, 1974) p. 163.

[5] N.C. Greenham, R.H. Friend and D.D.C. Bradley, Adv. Mater. 6 (1994) 491.

[6] J. Stampfl, S. Tasch, G. Leising and U. Scherf, Synth. Metals, in press.

[7] I.D.W. Samuel, B. Crystal, G. Rumbles, P.L. Burn, A.B. Holmes and R.H. Friend, Chem. Phys. Letters 213 (1993) 472.

[8] L. Smilowitz, A. Hays, A.J. Heeger, G. Wang and J.E. Bowers, J. Chem. Phys. 98 (1993) 6504.

[9] S.J. Strickler and R.A. Berg, J. Chem. Phys. 37 (1962) 814.

[10] I.D.W. Samuel, B. Crystall, G. Rumbles, P.L. Burn, A.B. Holmes and R.H. Friend, Synth. Metals 54 (1993) 281.

[11] U. Lemmer, R.F. Mahrt, Y. Wada, A. Greiner, H. Bässler and E.O. Göbel, Appl. Phys. Letters 62 (1993) 2827.

[12] M. Yan, L.J. Rothberg, F. Papadimitrakopoulos, M.E. Galvin and T.M. Miller, Phys. Rev. Letters 73 (1994) 744.

[13] D. Braun, E.G.J. Staring, R.C.J.E. Demandt, G.L.J. Rikken, Y.A.R.R. Kessener and A.H.J. Venhuizen, Synth. Metals 66 (1994) 75.

[14] E.G.J. Staring, R.C.J.E. Demandt, D. Braun, G.J. Riken, Y.A.R.R. Kessener, T.H.J. Venhuizen, H. Wynberg, W. ten Hoeve and K.J. Spoelstra, Advan. Mater. 6 (1994) 934.

[15] J.W.T. Walsh, Photometry (Constable, London, 1953).
[16] P.L. Burn, D.D.C. Bradley, R.H. Friend, D.A. Halliday, A.B. Holmes, R.W. Jackson and A. Kraft, J. Chem. Soc. Perkin Trans. I (1992) 3225.
[17] F. Wudl and S. Hoger, PCT Patent Application WO 94/20589 (1991).
[18] M. Leclerc, F.M. Diaz and G. Wegner, Makromol. Chem. 190 (1989) 3105.
[19] R.D. McCullough, R.D. Lowe, M. Jayaraman and D.L. Anderson, J. Org. Chem. 58 (1993) 904.
[20] K.A. Murray, S.C. Moratti, D.R. Baigent, N.C. Greenham, K. Pichler, A.B. Holmes and R.H. Friend, Synth. Metals (1994), in press.
[21] N.C. Greenham, S.C. Moratti, D.D.C. Bradley, R.H. Friend and A.B. Holmes, Nature 365 (1993) 628.
[22] D.R. Baigent, F. Cacialli, R.H. Friend, N.C. Greenham, J. Gruner, A.B. Holmes and S.C. Moratti, Polym. Preprints, in press.
[23] K.S. Wong, D.D.C. Bradley, W. Hayes, J.F. Ryan, R.H. Friend, H. Lindenberger and S. Roth, J. Phys. C 20 (1987) L187.
[24] F. Papadimitrakopoulos, K. Konstadininis, T.M. Miller, R. Opila, E.A. Chandross and M.E. Galvin, Chem. Mater. 6 (1994) 1563.
[25] C.L. Gettinger, A.J. Heeger, J.M. Drake and D.J. Pine, J. Chem. Phys. 101 (1994) 1673.
[26] N.C. Greenham, A.R. Brown, D.D.C. Bradley and R.H. Friend, Synth. Metals 57 (1993) 4134.
[27] B. Xu and S. Holdcroft, Macromolecules 26 (1993) 4457.
[28] M. Furakawa, K. Mizuno, A. Matsui, S.D.D.V. Rughooputh and W.C. Walker, J. Phys. Soc. Japan 58 (1989) 2976.
[29] H. Antoniadis, L.J. Rothberg, F. Papadimitrakopoulos, M. Yan, M.E. Galvin and M.A. Abkowitz, Phys. Rev. B 50 (1994) 14911.
[30] I.D.W. Samuel, to be published (1995).
[31] J.H.F. Martens, D.A. Halliday, E.A. Marseglia, D.D.C. Bradley, R.H. Friend, P.L. Burn and A.B. Holmes, Synth. Metals 55 (1993) 434.
[32] G. Cnossen, K.E. Drabe and D.A. Wiersma, J. Chem. Phys. 98 (1993) 5276.