

**AFRL-ML-WP-TP-2007-533**

**HIGH INTENSITY LIGHT  
PROPAGATION IN InAs (Preprint)**

**Shekhar Guha, Leo Gonzalez, Srinivasan Krishnamurthy,  
and Zhi Gang Yu**



**JUNE 2006**

**Approved for public release; distribution unlimited.**

**STINFO COPY**

**The U.S. Government is joint author of this work and has the right to use, modify, reproduce, release, perform, display, or disclose the work.**

**MATERIALS AND MANUFACTURING DIRECTORATE  
AIR FORCE RESEARCH LABORATORY  
AIR FORCE MATERIEL COMMAND  
WRIGHT-PATTERSON AIR FORCE BASE, OH 45433-7750**

## NOTICE

Using Government drawings, specifications, or other data included in this document for any purpose other than Government procurement does not in any way obligate the U.S. Government. The fact that the Government formulated or supplied the drawings, specifications, or other data does not license the holder or any other person or corporation; or convey any rights or permission to manufacture, use, or sell any patented invention that may relate to them.

This report was cleared for public release by the Air Force Research Laboratory Wright Site (AFRL/WS) Public Affairs Office (PAO) and is releasable to the National Technical Information Service (NTIS). It will be available to the general public, including foreign nationals.

THIS TECHNICAL REPORT IS APPROVED FOR PUBLICATION.

*\*//signature//*

---

SHEKHAR GUHA, Ph.D.  
Agile IR Limiters  
Exploratory Development  
Hardened Materials Branch

*//signature//*

---

MARK S. FORTE, Acting Chief  
Hardened Materials Branch  
Survivability and Sensor Materials Division

*//signature//*

---

TIM J. SCHUMACHER, Chief  
Survivability and Sensor Materials Division

This report is published in the interest of scientific and technical information exchange and its publication does not constitute the Government's approval or disapproval of its ideas or findings.

\*Disseminated copies will show "*//signature//*" stamped or typed above the signature blocks.

<b>REPORT DOCUMENTATION PAGE</b>				<i>Form Approved</i> OMB No. 0704-0188	
<p>The public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Department of Defense, Washington Headquarters Services, Directorate for Information Operations and Reports (0704-0188), 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to any penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number. <b>PLEASE DO NOT RETURN YOUR FORM TO THE ABOVE ADDRESS.</b></p>					
<b>1. REPORT DATE (DD-MM-YY)</b> June 2006		<b>2. REPORT TYPE</b> Journal Article Preprint		<b>3. DATES COVERED (From - To)</b>	
<b>4. TITLE AND SUBTITLE</b>  HIGH INTENSITY LIGHT PROPAGATION IN InAs (Preprint)				<b>5a. CONTRACT NUMBER</b> IN-HOUSE	
				<b>5b. GRANT NUMBER</b>	
				<b>5c. PROGRAM ELEMENT NUMBER</b> 62102F	
<b>6. AUTHOR(S)</b> Shekhar Guha and Leo Gonzalez (Agile IR Limiters, Exploratory Development) Srinivasan Krishnamurthy and Zhi Gang Yu (SRI International)				<b>5d. PROJECT NUMBER</b> 4348	
				<b>5e. TASK NUMBER</b> RG	
				<b>5f. WORK UNIT NUMBER</b> M08R1000	
<b>7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)</b>  Agile IR Limiters, Exploratory Development Hardened Materials Branch, Survivability and Sensor Materials Division Materials and Manufacturing Directorate Air Force Research Laboratory Wright-Patterson Air Force Base, OH 45433-7750				<b>8. PERFORMING ORGANIZATION REPORT NUMBER</b>  AFRL-ML-WP-TP-2007-538	
- SRI International					
<b>9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)</b> Materials And Manufacturing Directorate Air Force Research Laboratory Air Force Materiel Command Wright-Patterson Air Force Base, OH 45433-7750				<b>10. SPONSORING/MONITORING AGENCY ACRONYM(S)</b> AFRL/MLPJ	
				<b>11. SPONSORING/MONITORING AGENCY REPORT NUMBER(S)</b> AFRL-ML-WP-TP-2007-538	
<b>12. DISTRIBUTION/AVAILABILITY STATEMENT</b> Approved for public release; distribution unlimited.					
<b>13. SUPPLEMENTARY NOTES</b> The U.S. Government is joint author of this work and has the right to use, modify, reproduce, release, perform, display, or disclose the work. PAO case number AFRL/WS 06-2024, 21 August 2006. Submitted to the Applied Physics Journal.					
<b>14. ABSTRACT</b> We present our experimental and theoretical results on nonlinear absorption of light in InAs. The nonlinear variation of output intensity as a function of input intensity and time are calculated by solving four coupled rate equations simultaneously. All required quantities, including two-photon absorption, free-carrier absorption, Auger and radiative recombination lifetimes, and intrinsic carrier densities, have been obtained from the underlying band structures. The calculated thickness and energy-dependent output intensities in InAs agree very well with the values measured in our pump-probe experiments.					
<b>15. SUBJECT TERMS</b> Nonlinear absorption (NLA), Two-photon absorption (TPA), Free-carrier absorption (FCA)					
<b>16. SECURITY CLASSIFICATION OF:</b>			<b>17. LIMITATION OF ABSTRACT:</b> SAR	<b>18. NUMBER OF PAGES</b> 14	<b>19a. NAME OF RESPONSIBLE PERSON (Monitor)</b> Shekhar Guha, Ph.D. <b>19b. TELEPHONE NUMBER (Include Area Code)</b> (937) 255-4588
<b>a. REPORT</b> Unclassified	<b>b. ABSTRACT</b> Unclassified	<b>c. THIS PAGE</b> Unclassified			

## High Intensity Light Propagation in InAs

Srinivasan Krishnamurthy,<sup>1</sup> Zhi Gang Yu,<sup>1</sup> Shekhar Guha,<sup>2</sup> and Leo Gonzalez<sup>2</sup>

<sup>1</sup>*SRI International, Menlo Park, CA 94025, USA*

<sup>2</sup>*Air Force Research Laboratory, Materials and  
Manufacturing Directorate, WPAFB, Ohio 45433*

(Dated: June 29, 2006)

## Abstract

We present our experimental and theoretical results on nonlinear absorption of light in InAs. The nonlinear variation of output intensity as a function of input intensity and time are calculated by solving four coupled rate equations simultaneously. All required quantities, including two-photon absorption, free-carrier absorption, Auger and radiative recombination lifetimes, and intrinsic carrier densities, have been obtained from the underlying bandstructures. The calculated thickness and energy-dependent output intensities in InAs agree very well with the values measured in our pump-probe experiments.

PACS numbers: PACS: 42.70.Nq, 42.70.Km, 42.87-d, 78.20.c

Nonlinear absorption in semiconductors continues to be of scientific and technological interest.<sup>1-4</sup> Technologically, nonlinear absorption (NLA) is being used in a number of applications, including frequency conversion, optical switches and optical limiting.<sup>1</sup> The importance of free carrier effects in controlling the infrared nonlinear optical properties has long been realized<sup>2-4</sup>. Consequently, considerable efforts were undertaken to understand the NLA and evaluate the parameters such as two-photon absorption (TPA) coefficient, free-carrier absorption (FCA) coefficient, refraction cross section, and lifetimes in semiconductors.<sup>5-10</sup> However, extracting parameters from the measured values usually requires careful design of experiments to isolate and remove the effect of other parameters on the observed results. Even when evaluated at appropriate limits, the extraction ignores the complicated interdependence of these parameters on temperature and carrier density. For example, TPA and FCA depend on carrier density, which in turn depends on temperature. Consequently, the extracted values differ substantially from each other.<sup>4-6,9,10</sup> In addition, it is not clear whether the values extracted in certain limits can be used in modeling of high-intensity light propagation where the photoexcited carrier distribution is far from equilibrium. There have also been several intuitive calculations to evaluate these parameters using either effective mass bands and  $\mathbf{k}\cdot\mathbf{p}$  bands with adjustable matrix elements.<sup>3,7-10</sup> The trends are often obtained correctly. However, for complete understanding and modeling of light propagation with improved predictability, an accurate evaluation of these parameters from full bandstructures needed to be carried out.

In this Letter, we report the results of transmission measurements carried out with (a)

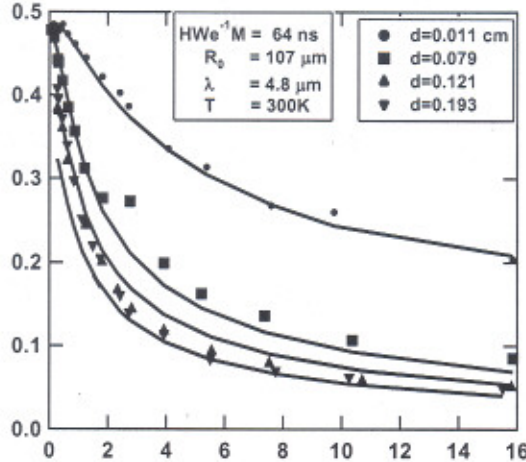


FIG. 1: Comparison of transmitted pump intensity measured (data points) at 300 K in  $10^{15} \text{ cm}^{-3}$  n-doped InAs with that calculated (solid lines) for four thicknesses.

pump only and (b) the pump-probe technique on a well-characterized InAs sample along with rigorous calculation of time ( $t$ ) and space ( $z$ ) dependent transmitted pump and probe intensity. All physical quantities including TPA and FCA coefficients, Auger recombination (AR) and radiative recombination (RR) lifetimes are calculated as functions of temperature ( $T$ ) and carrier density ( $N$ ) using full bandstructures. The calculated value of the transmitted probe intensity at the exit surface explains the experimentally observed trend very well and an excellent quantitative agreement is obtained when a Shockley-Read-Hall (SRH) mechanism is also included.

In our pump-probe experiment, the charge carriers were generated using TPA. In addition to the convenient availability of below band gap lasers, the TPA used here ensures that the entire thickness of the sample is utilized for carrier generation at low incident irradiance. The  $4.8 \mu\text{m}$  pump source was obtained by frequency doubling a TEA  $\text{CO}_2$  laser to provide 128 ns (full-width at  $e^{-1}$  of the maximum) duration pulses with energies up to 10 mJ. First, we carried out a set of NLA experiments at room temperature with only the pump present.

In Figure 1, the intensity transmission of the sample is plotted as a function of incident intensity for four different InAs sample thicknesses ( $d= 0.11 \text{ mm}$ ,  $0.79 \text{ mm}$ ,  $1.21 \text{ mm}$ , and  $1.93 \text{ mm}$ ). With increasing intensity, more carriers are created by TPA, and the resulting FCA decreases the output intensity nonlinearly with input intensity. As the thickness of the sample is increased, the path length of the light is longer and consequently more NLA takes

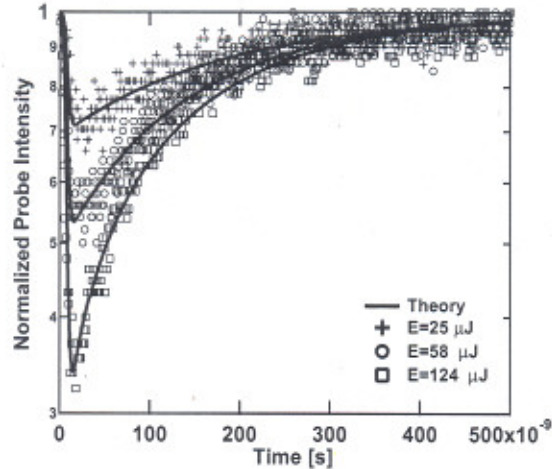


FIG. 2: Comparison of transmission intensity measured (data points) at 300 K in  $10^{15} \text{ cm}^{-3}$  n-doped InAs with that calculated (solid lines) for three beam energies.

place, as shown in Figure 1.

The decay time of the TPA excited carriers is studied using a pump-probe method. The pump is the  $4.8 \mu\text{m}$  beam described above and the probe is the *cw* laser beam obtained by a grating tuned CO laser operating at  $5.3 \mu\text{m}$  with power of 100 mW. The probe beam was chopped by a mechanical shutter to a 5 ms duration pulse. A dichroic filter (coated for high reflection at  $4.8 \mu\text{m}$  and anti-reflection at  $5.3 \mu\text{m}$  for 45 degree angle of incidence) was used to combine the pump and probe beams which were focused on the sample with the same 200 mm focal length lens to spot sizes of  $378 \mu\text{m}$  and  $166 \mu\text{m}$ , respectively (beam radius at  $e^{-1}$  of maximum). The sample studied is a 1-mm-thick InAs wafer with anti-reflection coating. The probe transmission was measured using a HgZnCdTe detector with a rise time less than 1 ns, and the incident and transmitted pump energies were measured using pyroelectric detectors.

Figure 2 displays the time dependence of the transmitted probe intensity (data points) through the sample for three different pump beam energies. At the onset of the pump pulse ( $t=0$ ), TPA in the material generates electron-hole pairs. Thus created free carriers absorb probe beam, by FCA, and cause a decrease in probe transmission. The transmission is minimum at the peak of the pump pulse. As the free carriers recombine, their density decreases and the FCA decreases. Consequently, the probe transmission increases to eventually reach its initial value. With an increase in the pump energy, from  $25 \mu\text{J}$  to  $124 \mu\text{J}$ , more carriers

are created at the end of the pulse and, consequently, the minimum in probe transmission is reduced further.

For a detailed understanding of the mechanisms affecting the light propagation in the material, a complete solution to the following rate equations is needed.

$$\frac{dI_P}{dz} = -\alpha I_P - \beta I_P^2 - \sigma(N_0 + N)I_P, \quad (1)$$

$$\frac{dN}{dt} = \frac{\beta I_P^2}{2h\nu} - \frac{N}{\tau_R} \quad (2)$$

$$\frac{dT}{dt} = \frac{\sigma^*(N_0 + N)I_P}{c_v} + \frac{\beta I_P^2}{c_v} \quad (3)$$

$$\frac{dI_{pr}}{dz} = -\sigma(N_0 + N)I_{pr}, \quad (4)$$

The equations relate the pump and probe intensities,  $I_P$  and  $I_{pr}$ , the concentration of the generated charge carriers  $N$ , and  $T$ .  $\alpha$ , and  $\beta$  are the one- and two-photon absorption coefficients, respectively, and  $\sigma$  ( $\sigma^*$ ) denote the sum of (difference in) the cross sections for FCA arising from photon emission and absorption. The total recombination rate  $\tau_R^{-1}$  is the sum of the AR rate ( $\tau_{ar}^{-1}$ ), RR rate ( $\tau_{rr}^{-1}$ ), and SRH rate ( $\tau_{srh}^{-1}$ ).  $N_0$  is the intrinsic carrier density at a given  $T$ . We have considered  $T$  increase due to the FCA and TPA, which has not been included in previous studies. We find that the increase in  $T$  can be significant (as much as 150 K for moderate intensities). It is important to realize that  $N_0$ ,  $\alpha$ ,  $\beta$ ,  $\sigma$ , and  $\tau_R$  are  $T$ -dependent. In addition,  $\beta$ ,  $\tau_R$  are dependent on  $N$ , which in turn depends on  $T$ . Moreover, the carrier density created by the pump beam affects the absorption of the probe beam. Hence, Eq. 1 through 4 have to be solved self-consistently. For the chosen value of incident pump intensity  $I_P(t=0, r, z=0)$  at a radial distance  $r$ , the time-dependent probe intensity  $I_{pr}(t, r, z)$  is evaluated at the exit side ( $z=L$ ) of the sample to compare with experiments. Since the photon energy,  $h\nu$ , is smaller than the bandgap of the materials considered,  $\alpha$  is zero. The self-consistent solution requires that  $N_0$ ,  $\beta$ ,  $\sigma$ , and  $\tau_R$  be calculated at all  $T$  and  $N$  values of interest. We calculate them in the range  $-10^{15} \text{ cm}^{-3} \leq N \leq 10^{18} \text{ cm}^{-3}$  and  $80 \text{ K} \leq T \leq 500 \text{ K}$ .

The TPA and FCA coefficients, RR and AR lifetimes, Fermi functions and intrinsic carrier concentrations are calculated using a hybrid pseudopotential tight-binding Hamiltonian that includes all long-range interactions.<sup>11</sup> In general, the  $\beta$  is assumed to be  $N$  and  $T$  dependent. However, we see that the Moss-Bernstein effect offers serious limitations to  $\beta$  at moderate

concentrations and low temperatures. For a given value of (equal electron and hole density)  $N$  and  $T$ , we first obtain the quasi-Fermi level, as appropriate for the non-equilibrium studies considered here. Then, using appropriate electron and hole distributions (for final and initial states, respectively),  $\beta$  is calculated.<sup>12</sup> We find that  $\beta$  (at  $4.8 \mu\text{m}$ ) remains nearly constant ( $0.45 \text{ cm/MW}$  for InAs) for all  $N$  until approximately  $10^{17} \text{ cm}^{-3}$  is reached, after which it decreases like a Fermi function for the given  $T$  and could be fitted very well to a functional form of  $\beta = a + be^{-c(T)n}$ , where  $n$  is carrier density in the unit of  $10^{-15} \text{ cm}^{-3}$ . Of the parameters ( $a, b, c$ ), we found only  $c$  depends on  $T$ .  $N_0$ , which is normally assumed to be constant,<sup>8</sup> is found to change by two orders of magnitude when the  $T$  is changed from 80K to 500K.

The FCA coefficient evaluated with full bands<sup>12</sup> is found to be proportional to  $N$ , in agreement with that assumed in the literature.<sup>8</sup> However, the proportionality constant,  $\sigma$ , which is the FCA cross section, is found to be strongly  $T$ -dependent. The contribution to the FCA comes more from holes than from electrons. The transition from light-hole to heavy-hole band does not require phonons, whereas the absorption of light by electrons is possible only with phonons. The  $T$ -dependence of phonon and electron distribution results in strongly  $T$ -dependent FCA cross section.

We modified the previously developed<sup>13</sup> full-bandstructure calculation of the lifetimes associated with electron-hole recombination through Auger and radiative processes to address the issue of electron and hole quasi Fermi levels. The calculated values of the lifetimes could be fitted accurately to a function form,  $\tau \equiv G(T)N^{-p(T)}$ .

All  $T$ -dependent parameters ( $N, c, G, p, \sigma$ ) are fitted accurately to a 4th order polynomial in  $T$ , for easier and more efficient use in obtaining self-consistent solutions to rate equations. The details of carrier density, coefficient, and recombination rate variation with  $T$  and  $N$ , and the fitted parameters, will be given elsewhere. Here we present only the transmission results obtained using these calculated values and parameters.

The calculated values of  $\beta(N, T)$ ,  $\sigma(T)$ , and  $\tau_R(N, T)$  are substituted in Eqs. 1-4 and solved for  $I_{pr}(z, r, t)$  and  $N(z, r, t)$  by the finite difference method with the boundary conditions  $I_P(0, r, t) = I_P^0 e^{-\left(\frac{r-t_0}{r_0}\right)^2} e^{-\left(\frac{r}{r_P}\right)^2}$ ,  $N(z, r, 0) = 0$ ,  $T(z, r, 0) = T_{Lat}$ , and  $I_{pr}(0, r, t) = I_{pr}^0 e^{-\left(\frac{r}{r_{pr}}\right)^2}$ , where  $r_P$  and  $r_{pr}$  are the pump and probe radii. The beam is assumed to be radially symmetric, and the beam energy,  $E$ , is the time and  $r$ -integrated value of  $I_P(0, r, t)$ . Care is taken to yield a numerically stable solution even under illumination of a strong pump laser

pulse. For a given incident pump irradiance, we obtain the value of the probe transmission as a function of  $r$ ,  $t$  and  $z$  in the sample. Then the transmission intensity is integrated over  $r$  to obtain a value at a given  $z$ . The transmission intensity evaluated at the exit surface ( $z=L$ ) is compared with experimental transmission curve. Although the calculated curve has trends similar to that observed, we find that inclusion of the SRH mechanism is needed for quantitative comparison with experiment. The SRH scattering is not intrinsic to the material and depends on external variables such as growth temperature and pressure. Our detailed calculations carried out previously<sup>14</sup> indicate a nearly T-independent SRH lifetime of 200 ns in  $2 \times 10^{16} \text{ cm}^{-3}$  doped InAs. Although the InAs sample studied here is only  $10^{15} \text{ cm}^{-3}$  n-doped, the excess carriers generated by the pump is of the order of  $1-2 \times 10^{16} \text{ cm}^{-3}$ , and justifies the use of the above calculated SRH lifetimes. We further assume that SRH lifetime decreases linearly with excess carrier density and obtain  $I_{pr}(L,t)$  in InAs for three values of  $E$ . The substrate ambient temperature 300K is used.

First, the equations are solved only for transmitted pump intensity without any probe present. We see in Fig. 1 that the calculated values (solid lines) as a function of input intensity agree very well with the values measured for four different InAs thicknesses. Then, for a fixed InAs thickness and three different beam energies, the transmitted probe intensity is calculated and compared with experiment, as shown in Fig. 2. The calculations explain the observed variations very well. It is important to note that good agreement is obtained for the pump transmission, probe transmission recovery, and the minimum of the probe transmission, which depend critically on all quantities: the intrinsic carrier density,  $\beta$ ,  $\sigma$ , temperature and the lifetimes. Only with N-dependence of SRH, included here in an approximation, an excellent agreement with experiment is obtained. The slight disagreement at large  $t$  is possibly because of actual N-dependence of SRH lifetimes, not considered here.

In conclusion, we have used pump-probe experiments to understand the optical interaction in InAs. The results are explained with accurately calculated TPA, FCA, AR, and RR lifetimes, and iterative solutions to coupled rate equations. Our calculations explain the observed variation in output pump intensity and time- and energy-dependent probe intensity accurately. We showed that with accurate evaluation of all parameters, the modeling can provide detailed guidance in understanding the underlying mechanisms and in choosing materials for light propagation applications.

SK and ZY gratefully acknowledge the funding from WPAFB contract F33615-97-D-5403

through Anteon Corporation.

---

- <sup>1</sup> For a review, E. Garmire, *IEEE J. on Selected Topics in Q. Elec.*, **6** 1094 (2000) and the references cited therein.
- <sup>2</sup> B. Jenson, *Infrared and millimeter waves*, Ed. K. Button (Academic, New York, 1982), Vol. 8.
- <sup>3</sup> E. van Stryland, Y.Y. Wu, D.J. Hagen, M.J. Soileau, and K. Mansour, *Opt. Soc. Am.*, **B5**, 1980 (1988).
- <sup>4</sup> M.B. Haeri, S.R. Kingham, and P. Milsom, *J. Appl. Phys.* **99**, 13514 (2006).
- <sup>5</sup> K.W. Berryman and C.W. Rella, *Phys. Rev.* **B55**, 7148 (1997).
- <sup>6</sup> B.N. Murdin, C.R. Pidgeon, A.K. Kar, D.A. Jaroszynski, J.-M. Ortega, R. Prazeres, and F. Glotin, *Opt. Mater.* **2**, 89 (1993).
- <sup>7</sup> M.S. Bahaei, D.C. Hutchings, D.J. Hagen, and E. Van Stryland, *IEEE J. of Q. Elec.*, **27**, 1296 (1991).
- <sup>8</sup> A. Kobayakov, D.J. Hagan, and E.W. Van Stryland, *J. Opt. Soc. Am.* **B 17**, 1884 (2000).
- <sup>9</sup> A. Miller, A. Johnson, J. Dempsey, J. Smith, C.R. Pidgeon and G.D. Holah, *J. Phys. C* **12**, 4839 (1979).
- <sup>10</sup> M.S. Bahaei, P. Mukherjee, and H.S. Kwok, *Opt. Soc. Am.* **B3** 379 (1986); and references cited there in.
- <sup>11</sup> A.-B. Chen and A. Sher, *Semiconductor Alloys*, (Plenum, New York, 1995).
- <sup>12</sup> S. Krishnamurthy, A. Sher, and A.-B. Chen, *J. Appl. Phys.* **88** 260 (2000).
- <sup>13</sup> S. Krishnamurthy, A. Sher, and A.-B. Chen, *J. Appl. Phys.* **82** 5540 (1997).
- <sup>14</sup> S. Krishnamurthy and M. Berding, *J. Appl. Phys.*, **90**, 848 (2001)