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Galvanomagnetic Luminescence of Indium Antimonide*

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Abstract

We report measurements of the absolute spectral intensity due to galvanomagnetic luminescence of intrinsic InSb at room temperature. Together with a calculation of carrier and photon transport, these measurements form the basis for a new technique for the determination of carrier lifetimes and diffusion lengths. The spectrally integrated luminescence has a bilinear form in terms of the exciting current density j and magnetic field B :

$$\Delta F = G \ B \ j \ ,$$

where ΔF is the change in the total emitted radiant energy flux from the thermal equilibrium value ($j = 0$). In particular ΔF can be negative. The new thermodynamic transport coefficient G has the value 1.8 μ W A^{-1} T^{-1} at 33°C, and decreases with increasing temperature.

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The phenomenon of galvanomagnetic luminescence (GML) is the modulation of ordinary thermal emission by the application of mutually orthogonal electric and magnetic fields, parallel to an emitting surface. The part of the equilibrium thermal emission which is due to electron-hole recombination is modulated as the carrier concentrations near a crystal surface are altered by the electric and magnetic fields. Based on thermodynamic considerations, for small current density $j_x > 0$ in the x-direction and small magnetic field $B > 0$ in the z-direction, the Lorentz force on mobile charges is expected to increase the total radiant emission ΔF in the negative y-direction:

$$\Delta F = G \; B \; j_x \; . \tag{1}$$

Here ΔF has dimensions of $\mathrm{Wm^{-2}}$, where $G > 0$ is the GML coefficient.

Because ΔF can have either sign, and is *thermodynamically reversible* by a change in the sign of B or j_x, ΔF can be used to produce a heat pumping action, for cooling or heating, provided that the coefficient G is sufficiently large, and that eq.(1) applies over a wide enough range of B and j_x to make ΔF observable. As will be shown here, for intrinsic InSb at room temperature $G = 1.8\ \mu\mathrm{W\ A^{-1}\ T^{-1}}$ and thus with $B = 2$ tesla and $j_x = 100\ \mathrm{A\ cm^{-2}}$, the available cooling/heating rates are on the order of 4 $\mathrm{Wm^{-2}}$. While the available cooling rate is small, it is by no means negligible. Maximum available radiative heat pumping rates have been discussed in another publication.[1]

The first GML observations on InSb were made by Ivanov-Omskii, *et al.* in 1965.[2] The same group reported[3] low resolution spectral measurements on p-type materials under high excitation (smallest excitation was $j \approx 2000\ \mathrm{A\ cm^{-2}}$, $B = 0.5$ T). These observations established that the emission was band-to-band recombination, and that it was several times more intense for one polarity of the current compared to the other. Recently, Morimoto and Chiba have shown that the emission from intrinsic InSb is linear in current up to about $\pm$ 200 $\mathrm{A\ cm^{-2}}$ at 2 T,[4] and is approxi-

mately linear in magnetic field, at low currents, up to $\pm$ 2T. [5,6] Measurements by Bolgov *et al.*, [7,8] focusing on the "negative luminescence" aspect of GML from InSb, have shown evidence that the equilibrium thermal emission from InSb due to band-to-band recombination can be suppressed almost entirely by fields of 1 T and a few tens of volts per cm ($j \approx 10^4$ A cm^{-2}).

In the remainder of this paper we outline the theory of galvanomagnetic luminescence under the restriction of weak excitation ($\Delta n << n_o$, $\Delta p << p_o$, where Δn and Δp are the changes from the equilibrium electron and hole concentrations n_o and p_o) and compare it with our experimental results. This comparison yields an improved value for the room temperature recombination lifetime.

Consider the transport of electrons and holes in a crystal with its surface in the $y = 0$ plane, extending in the positive y-direction a distance large compared to carrier diffusion lengths and the photon absorption length. With suitable electric and magnetic fields there is a drift of carriers in the negative y-direction due to the Lorentz force. These carriers accumulate near the free surface at $y = 0$ until they recombine radiatively or by some other mechanism. (Auger recombination is dominant at room temperature.) In a stationary state we expect

$$\Delta n(y) = \Delta n(0)\, e^{-y/L} , \tag{2}$$

where $\Delta n(y)$ is the excess electron concentration $\Delta n(y) = n(y) - n_o$, $\Delta p(y) = p(y) - p_o = \Delta n(y)$, and L is an effective ambipolar diffusion length. A knowledge of $\Delta n(y)$ will permit us to calculate the modulation of the radiative emission. Let J be the number of electron-hole pairs per unit time per unit area which are approaching the $y = 0$ surface. These pairs recombine at the rate $\Delta n(y)/\tau$ per unit volume in the interior, and at the rate $s\,\Delta n(0)$ per unit area at the surface. Here τ is the excess carrier lifetime and s is the surface recombination velocity parameter. An integration over y gives

$$J = (s + L/\tau)\Delta n(0) . \tag{3}$$

Under the assumptions of carrier non-degeneracy and the equality of drift and Hall mobilities it is not difficult to show (e.g., following Lile[9]) that

$$qJ = \left[\frac{\mu_e \mu_h (\mu_e + \mu_h) n_o p_o}{(n_o \mu_e + p_o \mu_h)^2 + (n_o - p_o)^2 \mu_e^2 \mu_h^2 B^2} \right] j_x B , \tag{4}$$

where q is the magnitude of the electron charge and μ_e , μ_h are electron and hole mobilities. This result shows that J is a non-linear function of B , except for the special case $n_o = p_o$ of intrinsic material. A quadratic equation for L can be obtained from Eq.(9) of Lile's paper.[9] For intrinsic material it reads:

$$\left(1 + \mu_e \mu_h B^2 \right) L^2 + 2\mu_e \mu_h \tau B E_x L - \frac{2\mu_e \mu_h}{\mu_e + \mu_h} \frac{kT}{q} \tau = 0, \tag{5}$$

where E_x is the electric field in the x -direction.

Consider now the photon propagation in the medium with the excess (or deficit) carrier spatial distribution given by (2). Radiant emission increases by the factor $np/n_o p_o$ compared to thermal equilibrium, while the absorptivity of the medium is decreased by this factor. Linearizing with respect to Δn and using $\Delta p = \Delta n$, one obtains the stationary transport equation for photons moving in the negative y direction, making an angle θ with the y-axis:

$$\frac{dR(\theta,y,\nu)}{dy} = \frac{\alpha(\nu)}{\cos\theta} \left[1 - \frac{n_o + p_o}{n_o p_o} \Delta n(y) \right] R(\theta,y,\nu) - \frac{\alpha(\nu)}{\cos\theta} N^2 b(\nu) \left[1 + \frac{n_o + p_o}{n_o p_o} \Delta n(y) \right] . \tag{6}$$

Here $R(\theta,y,\nu)$ is the energy transfer per unit of solid angle, of photon energy, and area, $\alpha(\nu)$ is the equilibrium photon absorptivity, N is the index of refraction (dispersion is neglected), and $b(\nu)$ is the Planck function per unit of solid angle, photon energy, and area:

$$b(\nu) = 2\nu^3 c^{-2} \left(\exp\left(\frac{h\nu}{kT} \right) - 1 \right)^{-1} . \tag{7}$$

In thermal equilibrium Eq.(6) reduces to $R\,(\theta,y\,,\nu) = N^2\ b\,(\nu)$, the well known result that black-body radiation in a medium of index N is a factor of N^2 more intense than in a vacuum. The modulated radiant energy transfer defined by

$$\Delta R\,(\theta,y\,,\nu) = R\,(\theta,y\,,\nu) - N^2 b\,(\nu)$$

is in linear approximation, using (2),

$$\Delta R\,(\theta,y\,,\nu) = \left(1 + \frac{\cos\theta}{\alpha(\nu)L}\right)^{-1} 2N^2 b\,(\nu) \left(\frac{n_o + p_o}{n_o\,p_o}\right) \Delta n\,(0)\ e^{-y/L}\ . \tag{8}$$

The GML energy flux leaving the medium, per unit photon energy and area, is

$$\Delta F\,(\nu) = 2\pi \int d\cos\theta\ \cos\theta\ \Delta R\,(\theta,y=0,\nu)\ T\,(\theta) \tag{9}$$

where the transmission of the interface $T\,(\theta)$ is $4N\,(N+1)^{-2}$ at θ=0 and falls to zero at $\theta = \arcsin N^{-1}$. A good approximation for $n = 4$ is to merely evaluate the integrand at θ=0 and take $\arcsin N^{-1} \approx N^{-1}$. (Error is $\leq 5\%$[10].) Collecting results now, we have, from Eqs. 3, 4, 8 and 9, for the intrinsic case,

$$\Delta F\,(\nu) = \frac{16N}{(N+1)^2} \left(\frac{\pi b\,(\nu)}{L + \alpha^{-1}(\nu)}\right) \left(\frac{s}{L} + \tau^{-1}\right)^{-1} \left(\frac{\mu_e\,\mu_h\,B}{\mu_e + \mu_h}\right) \left(\frac{j_x}{q n_o}\right) . \tag{10}$$

This result indicates that $\Delta F\,(\nu)$ is indeed proportional to $B j_x$. However, a non-linear dependence on B and E_x can occur in L . See Eq.(5). The integrated flux is

$$\Delta F = h \int_0^\infty d\,\nu\ \Delta F\,(\nu) \tag{11}$$

The InSb samples had donor concentrations and electron mobilities of $\approx 3 \times 10^{14}$ cm^{-3} and $\approx 5 \times 10^5$ cm^2 V^{-1} sec^{-1} at 77K. They were etched with either CP-4 or a dilute solution of bromine in methanol, and mounted on a copper heat sink with double-sided tape. Their nominal dimensions were 4 x 6 x 0.4 mm^3. Electrical leads were attached by ultrasonic soldering with indium. The magnetic field was applied continuously, and the electric field was modulated sinusoidally at 2 kHz. The emitted radiation was focused on the entrance slit of a grating monochro-

mator with a liquid nitrogen cooled Hg_{1-x} Cd_x Te photoconductive detector at the output slit. A preamplifier and lock-in amplifier completed the electronics. The optical system was calibrated by replacing the sample by a small heated blackbody source with a mechanical chopper, operating at 100 Hz.

Our measured data is shown in Fig. 1. The continuous theoretical curve is based on Eq. (10), with τ and L used as fitting parameters. The other parametric values employed are (for $T = 33^o$ C) $N = 4.0$, $s = 0$ (discussed below), $\mu_e = 7.27$ x 10^4 cm^2 V^{-1} sec^{-1}, $\mu_h = 766$ cm^2 V^{-1}sec^{-1}, $n_o = 2.2$ x 10^{16} cm^{-3}, $q = 1.602$ x 10^{-19}C. The data for $\alpha(\nu)$ were obtained from Ref. 11, and are for 22°C. At low photon energies the data fall above the fitted curve, due to the 3 meV decrease in the bandgap caused by sample heating to 33°C.[12] A smaller shift of the bandgap, to larger values, occurs due to the application of a magnetic field.[13] The structure in the data near the peak is an artifact due to water vapor in the optical path; more vapor was present during the spectral measurement than was present during the blackbody calibration. The fitting parameters τ and L were obtained by requiring that the computed peak height have the same value as the data, and that the computed curve agree with the data at 250 meV (where water vapor absorption is small). We obtain the lifetime $\tau = 6.0 \pm 1.2$ nsec and the effective diffusion length (at $B = 1.9$ T) $L = 3.0 \pm 1.0$ μm. The primary uncertainty in the lifetime is due to the $\approx$10% probable error in the absolute radiometric calibration. The primary uncertainty in L is probably errors in $\alpha^{-1}(\nu)$. If the value we have used, $\alpha^{-1}(250 \text{ meV}) = 4.0\mu m$, is in error by 10%, then the value of L is in error by 30%. A check on the consistency of the measurement can be made by computing L as a function of B , using the measured value of τ in Eq.(5). The term proportional to BE_x in (5) may be neglected because the driving electrical field oscillates about zero. We obtain 4.9 μm at B =0 and the value 2.8 μm at B =1.9T in agreement with the direct

measurement. Thus the separate measurements of τ and L are consistent.

The few published measurements of τ at room temperature, based on the photoelectromagnetic (PEM) effect, photoconductivity (PC) measurements, or photoluminescence measurements,[14,15,16] show scatter but are generally larger than our value. For example, Zitter, Straus and Attard[14] show values between 18 and 30 nsec based on PEM measurements, and larger values still for PC measurements. Recent photoconductivity kinetics measurements at Kiev,[17] however, show that τ does not exceed 30 nsec. For PEM and PC measurements on high mobility InSb films of thickness 0.8 to 30 μm, Hanus and Oswaldowski,[18] found they had to assume a value as small as τ=10 nsec, corresponding to L =5.5 μm (for μ_h = 600 cm^2 V^{-1} sec^{-1}), in order to fit their data. Consequently, their work lends support to our determination that the lifetime is only 6 nsec. *Ab initio* calculations of Auger lifetimes usually yield only order-of magnitude estimates; therefore it is notable that Gel'mont[20] has obtained a theoretical value of 10 nsec. While we believe we have observed the intrinsic lifetime, we cannot entirely rule out the possibility that crystalline imperfections have reduced the lifetime. More comprehensive studies, currently underway, should resolve this issue and also determine the doping dependence of the lifetime.

It has been assumed that the surface recombination velocity is small, i.e., that $s << L/\tau = 500$ m sec.$^{-1}$ This is consistent with observations by others for etched surfaces[9,17,19] and is also consistent with the fact that the intensity we measure is identical (to $< 10\%$) for samples etched with different chemicals. A sample with a surface polished with a fine alumina abrasive gave an identical spectrum as shown in Fig. 1, but with about 1/6 the intensity. This behavior is in agreement with Eq. 10 and gives a value of $s \approx 2500$ m sec^{-1} for a polished surface.

Integration over the spectrum of Fig.(1) gives the value $\Delta F = 3.47$ Wm^{-2}, and, with Eq.(1),

the coefficient of galvanomagnetic luminescence is $G = 1.8\ \mu W\ A^{-1}\ T^{-1}$. Based on further observations (at $\lambda = 5.5\ \mu m$), the temperature coefficient of G at room temperature is roughly -0.7% per degree C. This negative temperature sensitivity is to be expected because the Auger recombination which dominates the overall lifetime τ makes τ a stronger decreasing function of temperature than the radiative lifetime. Morimoto and Chiba[4] report approximate GML values for InSb $\approx 640\ \mu W\ cm^{-2}$ at $j = 14.3\ A\ cm^{-2}$ and $B = 2T$. This estimate gives $G \approx 22\ \mu W\ A^{-1}\ T^{-1}$, an order of magnitude larger than our value for G. The same paper (their Fig.3), however, shows that the linearity of the "negative" or suppressed luminescence extends to about 200 A cm^{-2} at 2T, implying that $\Delta F = 8.8\ mW\ cm^{-2}$. However, the equilibrium thermal emission of InSb for wavelengths less than 7.5 μm at 300K is easily computed to be about 3.5 mW cm^{-2}. Thus we believe that their (implicit) value for G is in error, since the absolute emission must remain positive.

In conclusion, we have demonstrated a new technique for the measurement of carrier lifetimes in narrow bandgap semiconductors. Since the radiative lifetimes are easily obtained from optical absorption measurements with the van Roosbroeck-Shockley relation, the quantum efficiency for conversion of excess electron-hole pairs to photons can be determined. For InSb at 33°C the radiative lifetime is about 580 nsec,[10] so our measured value $\tau = 6.0$ nsec implies that the quantum efficiency is 1.0%.

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References and Footnotes

1. P. Berdahl, *J. Appl. Phys.*, 1 August 1985. Ms No. R-6895.

2. V.I. Ivanov-Omskii, B.T. Kolomiets, and V.A. Smirnov, *Sov. Phys. Doklady 10,* 345-346 (1965).

3. V.I. Ivanov-Omskii, B.T. Kolomiets, and V.A. Smirnov, *Sov. Phys. J.E.T.P. Lett. 3,* 185-187 (1966).

4. T. Morimoto and M. Chiba, *Jap. J. Appl. Phys. 23,* L821-L823 (1984).

5. T. Morimoto and M. Chiba, *Phys. Lett. 85A,* 395-398 (1981).

6. T. Morimoto and M. Chiba, *Phys. Lett. 95A,* 343-344 (1983).

7. S.S. Bolgov, V.K. Malyutenko, and V.I. Pipa, *Sov. Tech. Phys. Lett. 5,* 610-611 (1979).

8. S.S. Bolgov, V.K. Malyutenko, and V.I. Pipa, *Sov. Phys. Semicond. 17,* 134-137 (1983).

9. D.L. Lile, *Phys. Rev. B 8,* 4708-4722 (1973).

10. T.S. Moss, G.J. Burrell, and B. Ellis, *Semiconductor Opto-Electronics,* John Wiley (New York), (1973).

11. T.S. Moss, S.D. Smith, and T.D.F. Hawkins, *Proc. Phys. Soc. (London) B70,* 776-784 (1957).

12. J. Camassel and D. Auvergne, *Phys. Rev. B 12,* 3258-3267 (1974).

13. E. Burstein, G.S. Picus, H.A. Gebbie and F. Blatt, *Phys. Rev. 103,* 826-828 (1956).

14. R.N. Zitter, A.J. Strauss, and A.E. Attard, *Phys. Rev. 115,* 266-273 (1959).

15. S.W. Kurnick and R.N. Zitter, *J. of Appl. Phys. 27,* 278-285 (1956).

16. J. Pehek and H. Levinstein, *Phys. Rev. 140,* A576-A586 (1965).

17. V.K. Malyutenko, S.S. Bolgov, V.I. Pipa and V.I. Chalkin, *Sov. Phys. Semicond. 14,* 457-460 (1980).

18. W. Hanus and M. Oszwaldowski, *Phys. Stat. Sol. 36,* 445-452 (1976).

19. H. Fujisada, *J. Appl. Phys. 45,* 3530-3540 (1974).

20. B.L. Gel'mont, *Sov. Phys. JETP 48,* 268-272 (1978).

Figure Caption

Fig. 1. Measured and computed spectra of galvanomagnetic luminescence from intrinsic InSb.

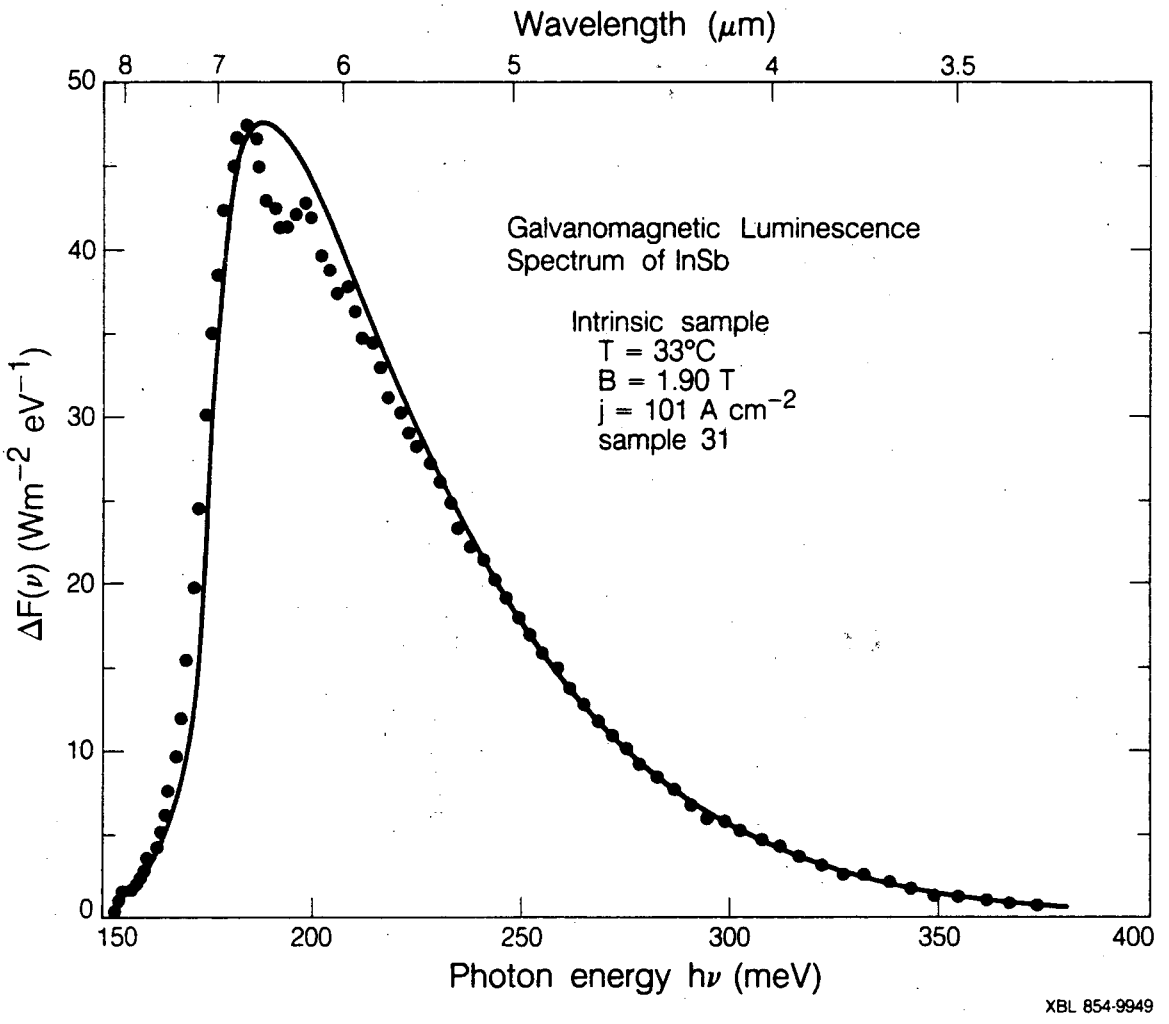
Wavelength (μm)
8
7
6
5
4
3.5
50
40
30
20
10
0
ΔF(ν) (Wm⁻² eV⁻¹)
Galvanomagnetic Luminescence
Spectrum of InSb
Intrinsic sample
T = 33°C
B = 1.90 T
j = 101 A cm⁻²
sample 31
150
200
250
300
350
400
Photon energy hν (meV)
XBL 854-9949

Figure 1.

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