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MAGNETIC FIELD DEPENDENCE OF LASER EMISSION IN $Pb_{1-x}Sn_xSe$ DIODES*

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The magnetic field dependence of long-wavelength infared laser emission has been studied in $Pb_{1-x}Sn_xSe$ diodes for compositions in the range $0 \leq x \leq 0.3$. For $x > 0.15$ the energy of the lowest transition decreases with increasing magnetic field whereas for $x < 0.15$ this energy increases. This unique observation is consistent with a theory of magnetic energy levels proposed by Baraff and also strongly supports the inversion model for the energy bands in Pb-Sn chalcogenides.

The temperature dependence of laser emission in $Pb_{1-x}Sn_xSe$ diodes in the composition range $0 \leq x \leq 0.3$ has recently been reported[1] and the magnetic field dependence of laser emission and spontaneous luminescence has been measured for PbS, PbSe, and PbTe[2] and for low-SnTe-content $Pb_{1-x}Sn_xTe$ diodes.[3] We report here the results of measurements made on a number of $Pb_{1-x}Sn_xSe$ diode lasers in the above composition range at magnetic fields up to 145 kG. The results support the model previously proposed[4] in which the valence- and conduction-band states in $Pb_{1-x}Sn_xSe$ alloys approach each other, invert, and move apart as SnSe is added to PbSe. At low temperatures this inversion occurs at about $x = 0.15$. For the alloys on the SnSe-rich side of the inversion point the lowest energy transition between magnetic levels of the conduction and valence bands has an energy which decreases with increasing magnetic field. To our knowledge this is the first such observation in any material. In addition the results give a direct indication of the effect that the higher lying energy bands have on the conduction and valence-band-edge masses and g factors in these materials.

For the magnetic-field measurements the diodes were near liquid-He temperature and oriented with the diode current parallel to the magnetic field in a $\langle 100 \rangle$ crystallographic direction. The laser radiation was emitted perpendicular to this direction. The photon energy of the laser emission as a function of magnetic field is shown in Fig. 1 for diodes of $Pb_{1-x}Sn_xSe$ with $x = 0$, 0.05, and 0.10 and in Fig. 2 for $x = 0.19$, 0.22, and 0.28. The Landau levels shown schematically in the insets identify the transitions observed. At low magnetic fields one generally observes the line T_2. As the field is increased the emission switches to T_1. If the diode current is increased the T_2 emission persists up to higher magnetic field values and in some cases a third line T_3 is observed. For all of the alloys studied with $x < 0.15$ (including PbSe) the T_1 line is found to have a positive slope, in most cases about 10^{-7} eV/G. For alloys with $x > 0.15$ the slope of this line is negative but has about the same magnitude. The zero-field energy gap as a function of alloy composition is shown in Fig. 3. The mole fraction of SnSe (x) was measured using an electron microprobe, and the energy-gap values were obtained from extrapolations of the magnet-

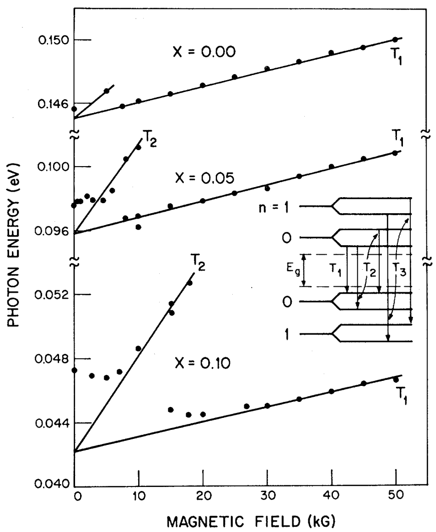

FIG. 1. Magnetic field dependence of laser emission in $Pb_{1-x}Sn_xSe$ diodes with $x=0.00$, 0.05, and 0.10. The data for PbSe, $x=0.00$, were taken from Ref. 2.

ic-field data to zero field as indicated in Figs. 1 and 2. The emission energy at zero magnetic field is generally 2-4 meV above this extrapolated value, probably due to band filling.

The magnetic field dependence of the emission energy can be understood in terms of a band model[4] proposed for the $Pb_{1-x}Sn_xTe$ and $Pb_{1-x}Sn_xSe$ alloys and a theory of magnetic energy levels proposed by Baraff.[5] Consider first a simple two-band model for the extremal valence and conduction bands in which the interactions with the higher lying energy bands are ignored. Under these circumstances the effective-mass tensors and g factors of the conduction and valence bands are equal. In addition Cohen and Blount[6] have shown that there is a relationship between the effective masses and g factors, such that the lowest magnetic level of the conduction band and highest magnetic level of the valence band should each have an energy which is independent of magnetic field. Although the arguments of Cohen and Blount as well as those of Baraff were developed for bismuth, they are based on considerations which apply equally well to the Pb and Pb-Sn salts. Consequently, in this simple two-band model the lowest energy transition T_1 should also have an energy which is independent of magnetic field. The slope of the T_1 line in Figs. 1 and 2 is therefore a direct indication of the effects of the higher lying energy bands on the extremal conduction and valence bands in PbSe and $Pb_{1-x}Sn_xSe$.

Simple arguments[7] as well as detailed calculations[8-10] indicate that there should be six energy bands in the vicinity of the energy gap at the L point in the lead salts. It is apparent from our data as well as those of others[11-13] that the other four bands must contribute significantly to the valence and conduction band masses and g factors. Baraff has extended the simple two-band model by including the interaction with the higher lying energy bands in second-order perturba-

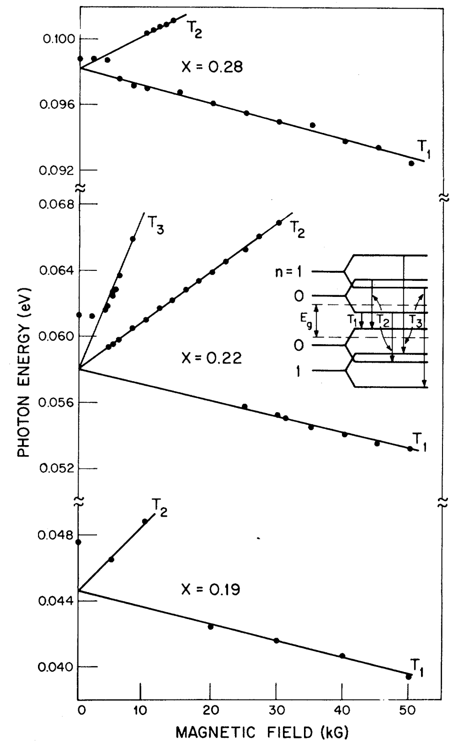

FIG. 2. Magnetic field dependence of laser emission in $Pb_{1-x}Sn_xSe$ diodes with $x=0.19$, 0.22, and 0.28.

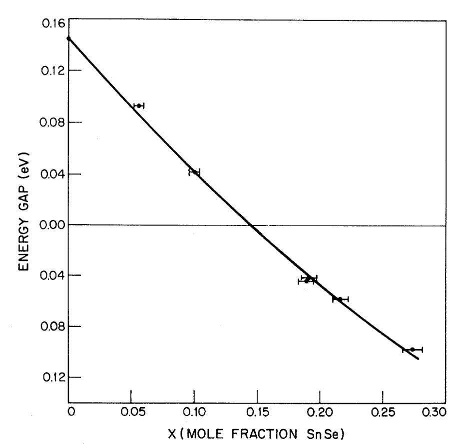

FIG. 3. Energy gap versus composition in $Pb_{1-x}Sn_x$-Se.

tion theory. This results, among other things, in a small linear field dependence of the lowest energy valence- and conduction-band magnetic levels, which in our case can be expressed as

$$E_{6^-} = E_g + (\tfrac{1}{2}A - B)H, \qquad (1)$$

and

$$E_{6^+} = +(\tfrac{1}{2}C - D)H, \qquad (2)$$

where the subscript 6^- refers to the L_6^- band which forms the conduction band in PbSe and 6^+ refers to the L_6^+ band which forms the valence band in PbSe,[9,10] and the energies are measured from the top of the valence band at zero field; A and C represent a deviation in the L_6^- and L_6^+ effective masses, respectively, and B and D represent a deviation in the L_6^- and L_6^+ g factors, respectively, from the values predicted by the two-band model.

The magnetic field dependence of the transition T_1 in Fig. 1 is about $+1.0\times10^{-7}$ eV/G for all three compositions which, from Eqs. (1) and (2), corresponds to $(\frac{1}{2}A-B)-(\frac{1}{2}C-D)$. According to the model[4] for the energy bands in the $Pb_{1-x}Sn_xSe$ alloys, for $x>0.15$ L_6^+ becomes the conduction band and L_6^- becomes the valence band. Neglecting any other changes, Eqs. (1) and (2) become in this case

$$E_{6^+} = E_g + (\tfrac{1}{2}C - D)H \qquad (3)$$

for the conduction band and

$$E_{6^-} = +(\tfrac{1}{2}A - B)H \qquad (4)$$

for the valence band, and the magnetic field dependence of the transition T_1 in these alloys should be given by $(\frac{1}{2}C-D)-(\frac{1}{2}A-B)$ which is just the negative of the field dependence in the alloys with $x<0.15$. The values of -0.97×10^{-7}, -0.96×10^{-7}, and -1.1×10^{-7} eV/G observed for $Pb_{0.81}Sn_{0.19}Se$, $Pb_{0.78}Sn_{0.22}Se$, and $Pb_{0.72}Sn_{0.28}Se$ diodes, respectively, are in good agreement with this prediction. The negative magnetic field dependence of the T_1 transition in all the alloys with $x>0.15$ so far examined lends additional support to the model involving the crossover of the L_6^- and L_6^+ states.

The fact that the absolute value of the slope of T_1 stays roughly constant with changing alloy composition from $x=0$ to $x=0.28$ indicates that the interactions of the L_6^+ and L_6^- bands with the other energy bands at the L point do not change much in this alloy range. This combined with the observation that the relative energies of the L_6^+ and L_6^- bands change by 0.24 eV in this range implies that the other energy bands are well separated from the conduction and valence bands. Energy-band calculations[9,10] indicate that this separation is of the order of 1-3 eV which is consistent with the above observations.

From the slope of T_1 we can obtain directly the quantity $2m_0/m_r^* - (g_c+g_v) \simeq 35$, where m_r^* is the reduced effective mass of the valence and conduction bands given by $1/m_r^* = 1/m_c^* + 1/m_v^*$ and m_v^*, m_c^*, g_v, and g_c are the $\langle 100\rangle$ band-edge effective masses and g factors of the valence and conduction bands, respectively. This quantity, which has the same magnitude for all alloy compositions, is equal to zero in the two-band model. Therefore it is a direct measurement of the interaction of the conduction and valence bands with the other bands at the L point.

As shown in Figs. 1 and 2, the next higher energy transitions following T_1 are the two transitions labeled T_2. No more than one T_2 line has been observed in any of the $Pb_{1-x}Sn_xSe$ diodes, and it has not been possible to determine if one transition or two equal-energy transitions are involved. If the transitions have equal energies the two transitions labeled T_3 also have equal energies and a magnetic field dependence three times that of the T_2 line. The highest energy emission line observed in $Pb_{0.78}Sn_{0.22}Se$ has approximately three times the slope of the T_2 line. We thus identify this line with the transition T_3. There are several allowed transitions with energies less than T_3. However, the probabilities for these transitions appear to be sufficiently smaller than those of T_3 and T_2 that their emission is not observed. Based on our identification of the T_1, T_2, and T_3 lines in $Pb_{0.78}Sn_{0.22}Se$, we can obtain $m_r^* = (0.018\pm0.001)m_0$, $|g_c - g_v| = 9\pm9$, and $|g_c+g_v| = 78\pm8$ for this composition.

For the other diodes studied, present data were insufficient for a reasonably accurate determination of both mass and g-factor values. Nevertheless, for alloys with the same absolute value of the energy gap, those on the SnSe-rich side of the band-inversion point appear to have a heavier effective mass than those on the PbSe-rich side. This can be understood qualitatively in terms of the positions of the other energy bands at the L point in PbSe.[9,10] The other energy bands which contribute significantly to the effective mass of the valence band all lie above the conduction band and those which contribute significantly to the effective mass of the conduction band all lie below the valence band, such that their effect is to decrease both the valence- and conduction-band masses from those predicted by the two-band model. However, when the conduction and valence bands invert, the effects of the other bands oppose those of the conduction and valence bands, such that the masses are heavier than predicted by the two-band model.

For small energy-gap alloys on the SnSe-rich side of the crossover, the negative magnetic field dependence of the laser-emission energy should enable the tuning of these lasers out to very long wavelengths. For $Pb_{0.81}Sn_{0.19}Se$, the T_1 line was observed up to 80 kG and 34 μ, which is the longest wavelength of semiconductor laser emission thus far obtained. However, the laser emission ceased at this point and was not observed at fields up to the maximum available field of 145 kG. For a second $Pb_{0.78}Sn_{0.22}Se$ diode, the T_1 line was observed to the maximum field, but at fields greater than 100 kG the magnetic field dependence deviated from a straight line with a diminishing slope. It should be noted that according to Baraff the energy of this lowest transition is linear with magnetic field, except that there are higher order terms which prevent the two lowest magnetic levels from crossing. However, we can estimate that the minimum separation energy is of the order of 10^{-4} eV at 100 kG if indeed the other energy bands are 1 to 3 eV from the conduction and valence bands. This is much too small to account for the deviation in slope for the $Pb_{0.78}Sn_{0.22}Se$ diode emission. The situation is more complicated, of course, if the other energy bands are near by.

A second possible cause for these effects arises from the fact that these diode lasers have bulk n- and p-type carrier concentrations in the 10^{17}- to 10^{18}-cm^{-3} range. The plasma energy for an alloy of composition $x = 0.19$ and a carrier concentration of 10^{18} cm^{-3} can be estimated to be approximately 0.034 eV or 37 μ which is in the energy range where these effects are occuring. It should also be pointed out that we expect the cyclotron resonance energy to be in this same range at 100 kG for these low-gap materials. One might expect an interaction with the plasma, magnetoplasma, or cyclotron-resonance modes which could affect the magnetic field dependence of the laser emission and possibly even the energy of emission at zero field. These possibilities are being studied further.

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