allowed two-photon transition from the crystal ground state. The dotted lines correspond to branches which cannot be reached by two-photon absorption.

In conclusion, we could, for the first time, give evidence of the biexciton dispersion. This dispersion is fully explained by a theoretical model taking into account $\vec{K}$-dependent interaction terms, already known from the exciton-photon problem.

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[a] Permanent address: Institut für Theoretische Physik, Universität Regensburg, D-8400 Regensburg, Germany.

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Photoexcited Exciton Spin-Flip Scattering in *p*-InSb

J. F. Scott
Department of Physics, University of Colorado, Boulder, Colorado 80309
(Received 15 October 1979)

Stimulated spin-flip scattering reported in p-InSb by Ebert *et al.* is reinterpreted as being due to excitons. The analysis explains the observation of Raman shifts which change magnitude as functions of pump wavelength at constant magnetic field. The Raman shifts satisfy the unusual equation $\Delta\omega = \pm \mu_B g^*(H - H_0)$, where $H_0 \cong 29$ kG, and g^* is about 6. The number of transitions (six) and their spacing as triplet, doublet, and singlet are explained by the model.

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In 1977 Ebert *et al.* published a remarkable set of data for spin-flip Raman scattering in p-type InSb.[1] To the author's knowledge, these data have never been explained, even qualitatively. The observed transitions exhibited frequency shifts which were linear in applied magnetic field, but three characteristics appeared unique and unexplainable: First, the frequency shifts, although linear in field, did not extrapolate to zero at zero field, but to zero frequency at large field values, of order 40 kG; second, the observed frequency shifts were different for each of six CO-laser pump wavelengths employed; third, for a given field value as many as three different transitions could be observed by changing the pump wavelength. Although Ebert *et al.* were unable to explain any of these three effects, they were certain that the observed transitions were indeed Raman scattering by virtue of the Stokes and anti-Stokes spectra observed in each case.

In this paper, I will reanalyze the data of Ref. 1 in terms of exciton transitions. It is found that a quantitative explanation of all of the data characteristics can be obtained in this fashion without the introduction of any adjustable parameters, merely with known values from the literature. There were two motivations for this exciton approach. Firstly, the early work on InSb lasers[2] demonstrated a key role for exciton states; and secondly, recent analysis[3] of spin-flip scattering in p-SiC revealed the presence of spin-flip scattering from photoexcited excitons. Both the electron g value ($g_e = 1.97$) and the electron-hole exchange energy ($\Delta_0 = 1.1$ meV) were obtained from an analysis of these data. Other photoexcited transitions have been measured in high magnetic fields in ZnTe:As.[4]

As early as 1963 Phelan *et al.* found[5] that diode or optically pumped[6] InSb samples emitted at energies 1 or 2 meV below the band gap in zero magnetic field; in addition, for small fields the output wave number varied quadratically with field, a second indication of excitons. In Ref. 5 it was suggested that the laser transitions observed corresponded to the transition from an upper state characterized by the $m=-\frac{1}{2}$ lowest Landau level to a lower state characterized by the $m=+\frac{1}{2}$ level of an electron bound to an impurity. This same kind of free-to-bound transition will be invoked for the Raman data of Ref. 1 in the present paper.

Figure 1 summarizes much of what is known concerning absorption and emission characteristics of InSb samples at liquid-helium temperatures. The open circles in this figure are absorption peaks due to excitons bound to impurities. The impurities were assigned as Zn or Cd on the basis of studies by Putley.[7] Johnson suggested that they are ionized,[8] but we know from Hopfield's work[9] that this is impossible and have assigned them as neutral. Excitons bound to neutral acceptors will have the spins of the exciton holes pair in an antiparallel fashion with the spins of the holes neutralizing the acceptors, leaving only the electron g value to produce doublet splitting in applied magnetic fields. This is shown in Fig. 1; note that g_e is negative, so the $M_s=-\frac{1}{2}$ state lies above the $+\frac{1}{2}$ state. By comparison, the free exciton will split into eight levels in applied fields, corresponding to M levels for the

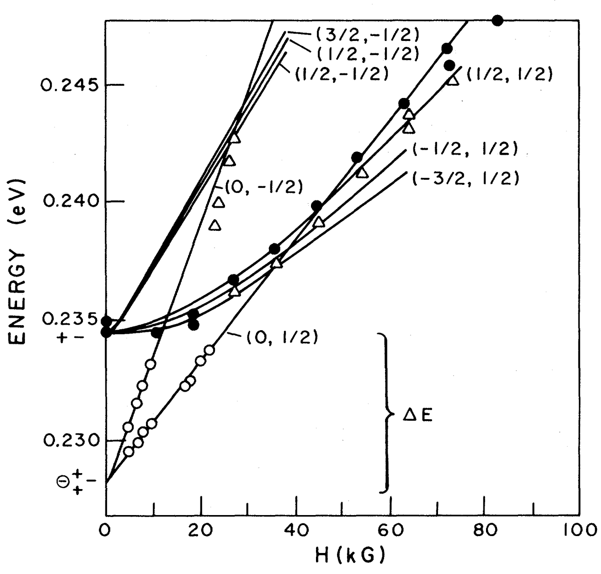

FIG. 1. Exciton energy vs magnetic field in p-InSb. Open circles are absorption maxima and originate at 0.229 eV, a level assigned as excitons bound to neutral acceptors (Zn or Cd). Triangles are stimulated emission energies and solid circles are spontaneous emission energies, both originating at the free exciton level 0.2347 eV at $H=0$.

$p_{3/2}$-like holes of $\pm\frac{3}{2}$, $\pm\frac{1}{2}$, and M levels of $\pm\frac{1}{2}$ for the electrons. States in Fig. 1 are labeled (M_{hole}, M_{electron}); the $M_J=\pm 2$ states are not shown, since they are not expected to be involved in the observed transitions. The level at ~0.235 eV agrees with Dimmock's calculation[11] for the free exciton. The solid curves in Fig. 1 are fits to a theory by Johnson and Fan[10] and are given by

$$\hbar\omega \cong [m_e/m_c^* \pm \tfrac{1}{2} g_e]\mu_B H + \hbar\omega_{\text{ex}}, \qquad (1)$$

where m_c^* is the conduction-electron effective mass of $0.015m_e$, and g_e is the conduction-electron gyromagnetic ratio ~51. The expression in Eq. (1) neglects the acceptor splittings and Coulomb behavior of the exciton, which produces a deviation from linearity at low values of H; it should be accurate for fields above 10 kG. The triangles are stimulated emission energies, and the solid circles are spontaneous-emission energies.[5] The Zeeman pattern is that predicted by the Kleiner-Roth Hamiltonian.[12]

Figure 2 diagrams the Raman transitions involved in the data of Ref. 1. Figure 2(a) shows the crossings of three of the free-electron levels: $(\frac{1}{2},\frac{1}{2})$, $(-\frac{1}{2},\frac{1}{2})$, and $(-\frac{3}{2},\frac{1}{2})$, with the $(0,\frac{1}{2})$ bound-electron state. In drawing Figs. 1 and 2 I have assumed that the hole gyromagnetic ratio in InSb is about the same as in GaP or ZnTe,[13,14] crystals with similar valence band structure. A best fit to the data of Ref. 1 yields $g_h=0.8$, which is the value shown in the figures.

The $M_J=\pm 1$ states in Fig. 2(a) do not have the same field dependence as does the $M_J=0$ state, leading to an uneven Zeeman splitting. Such M_J-dependent diamagnetic shifts are well known in other III-V semiconductors.[14-18]

Figure 2(b) presents a rather schematic diagram of the scattering process that I am invoking. The initial state is an electron bound to a hole (exciton); the intermediate state is a free-conduction electron; and the final state is an electron bound to two holes and a charged impurity (i.e., an exciton bound to a neutral acceptor). In this view the transitions observed are one-electron processes in which the local environment of the electron changes. Strong resonance effects are observed in Ref. 1, and we note that the six different CO-laser pump wavelengths used in Ref. 1 will resonate in turn with each of the six free-exciton levels shown in Fig. 1 at slightly different fields, from 30 to 55 kG. That is, the observation in Ref. 1 of six different stimulated transitions with six different pump wavelengths, heretofore unexplained, arises in my analysis as

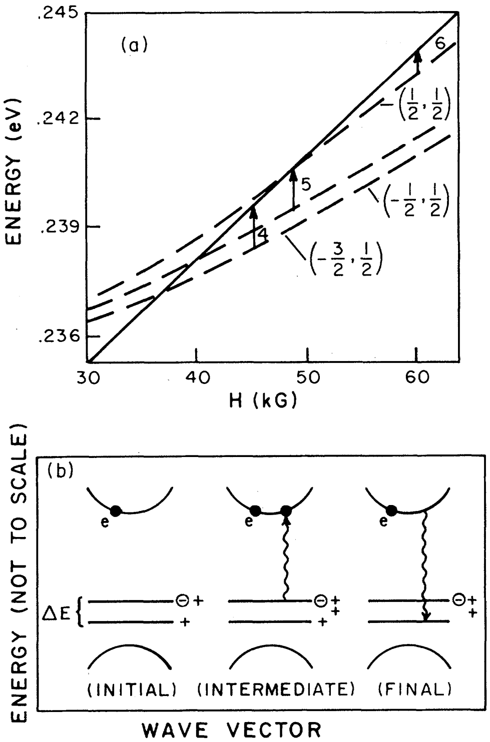

FIG. 2. (a) Exciton energies vs magnetic field showing crossing of levels near 30, 40, and 60 kG. (b) Schematic diagram of the exciton Raman-scattering process invoked for p-InSb. The horizontal lines show the hole-state energies. The initial state is a free exciton and a neutral acceptor. In the intermediate state the neutral acceptor traps a second hole. In the final state an electron annihilates the free-exciton hole.

due to the six free-exciton levels being tuned one at a time with applied magnetic field through the pump energies for energies for exact resonance.

Figure 3 reproduces the data of Ref. 1. The observed transitions agree in number (six) and field dependence with the diagrams in Figs. 1 and 2(a). Note that the zero-energy separations at about 30 to 40 kG are not adjustable; they are independent of the g_h value assumed in our analysis. In addition, the nearly linear but slightly sublinear field dependence of the observed Raman transitions is also in accord with Figs. 1 and 2(a) and arises from the different diamagnetic terms

$$E(H) - E(0) = (e^2a^2/4\mu c^2)H^2 \tag{2}$$

for the free and bound excitons, where a is the exciton Bohr radius and μ is its reduced mass.

The Raman transition energies satisfy Eq. (3) below:

$$\hbar\Delta\omega = \pm\mu_B g^*(H - H_0), \tag{3}$$

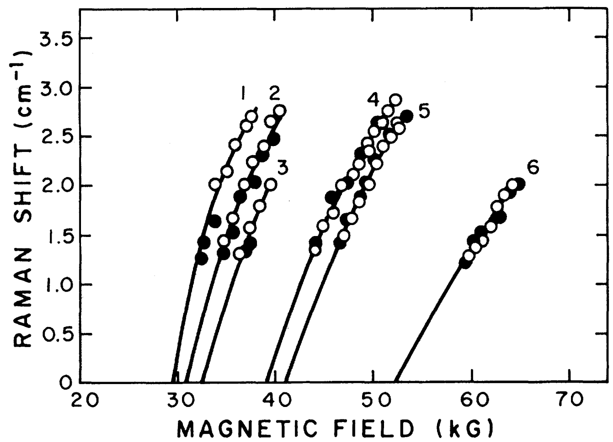

FIG. 3. Magnetic field dependence of the frequency shift of heavy hole Raman scattering at various pump frequencies. Solid circles have $p = 1.6\times 10^{13}$ cm^{-3}; open circles, $p = 1\times 10^{14}$ cm^{-3}. Curves 1–6 are for pump frequencies 1943.5, 1947.5, 1951.5, 1965.2, 1969.3, and 1995.1 cm^{-1}, respectively.

where H_0 is a complicated function of exciton binding energies and diamagnetic shifts, and g^* is an effective g value given by the difference in slopes of the free and bound exciton curves in Fig. 2(a) at their crossing field.

The parameter g^* in Eq. (2) is a function of the average slopes of the free-and bound-exciton levels at their crossings. This, in turn, derives from the diamagnetic, low-field term in Eq. (2).

Equation (2) can be used to estimate g^*. Alternatively, this can be done graphically from Fig. 2(a). The result is $g^* \simeq 6$. The expression for the observed transition frequencies is then

$$\hbar\omega_4 = (g^* + g_h)\mu_B(H - H_0), \tag{4}$$

$$\hbar\omega_5 = (g^*)\mu_B(H - H_0), \tag{5}$$

$$\hbar\omega_6 = (g^* - g_h)\mu_B(H - H_0), \tag{6}$$

with similar expressions involving a different g^* for transitions 1, 2, and 3 of Fig. 3. Equations (4)–(6) show that the spectra observed by Ebert *et al.*[1] are *not* really hole spin-flip scattering. Transitions 2 and 5 are free- to bound-exciton transitions in which neither g_e nor g_h play a direct role. All six transitions may be generally described as "hole unpairing" in magnetic fields, not as hole spin flip. In the initial-state exciton hole spin is aligned by the acceptor hole spin (antiparallel); in the final state it is aligned by the external magnetic field. Thus, there is more of a "hole spin flop," in analogy with magnetic insulators, than a hole "spin flip."

In summary, in this section I have explained the number (six) and spacing of the observed transitions.

The final aspect of the analysis concerns the temperature dependence and thresholds. Note the observation in Ref. 1; the transitions disappeared at temperatures above 2.5 °K. This in itself is compelling evidence that the states involved are excitonic. Why, however, does the free-electron spin flip disappear above 30 kG and the exciton process "turn on" at the threshold field? The probable answer is implicit in the diagram in Fig. 1. If we assume steady-state populations of exciton levels, and further assume that the Fermi level E_F in these very slightly p-type samples lies just below the bandgap, then both the impurity exciton levels and the free-exciton levels will be completely filled for fields less than about 30 kG. The Pauli exclusion principle excludes any Raman transitions between these states under such conditions. Above 30 kG, however, the bound exciton levels pop out above the free-exciton levels and also above E_F. Under these conditions a dramatic increase in transition probability suddenly occurs for the free- to bound-exciton transition. Its gain exceeds that of the conduction-electron spin-flip process, and the latter "turns off," as observed in Ref. 1.

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Unification, Monopoles, and Cosmology

J. N. Fry and David N. Schramm
Enrico Fermi Institute, The University of Chicago, Chicago, Illinois 60637
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It is shown that the cosmological problem of too many relic monopoles generated in the big bang according to standard grand unification theories can be avoided by several straightforward machanisms, one of which may make monopoles useful as seeds for galaxy formation.

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The apparent success of the Weinberg-Salam $SU(2)\otimes U(1)$ theory of weak and electromagnetic interactions[1] and the SU(3) color gauge theory of the strong interactions[2] leads irresista bly to grand unified theories[3-7] in which all interactions are contained in one large simple gauge group [the smallest containing $SU(3)\otimes SU(2)\otimes U(1)$ being[3] SU(5)], and where the apparent differences between forces observed today are the result of the breakdown of the exact symmetry at the low temperatures (compared to the unification scale) of the present universe. In addition to intrinsic

