Plasmaron Coupling and Laser Emission in Ag-Doped $CdSnP_2$

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We report the observation of laser emission in optically excited $CdSnP_2$:Ag at a threshold of ~50 kW/cm^2 at 20°K. From a study of the magnetic-field–induced shift in wavelength of the spontaneous and stimulated emission, the recombination mechanism is identified as band to band in a regime where electron–hybrid-plasmon (plasmaron) coupling is dominant. This is the first observation of plasmaron coupling in semiconductor laser action, and the first unambiguous experimental evidence for plasmaron coupling in semiconductors.

Although several II-IV-V_2 chalcopyrite semiconductors are known to have direct band gaps, the only report of laser emission in these crystals has been in nominally pure $CdSnP_2$ excited with an electron beam.[1] The threshold current density for laser emission using a 25-keV electron beam was surprisingly low at 0.2 A/cm^2, less than the threshold for GaAs lasers excited in a similar fashion. In the present Letter we report the observation of efficient spontaneous and stimulated emission in Ag-doped (~10^{18} cm^{-3}) $CdSnP_2$. In crystals cleaved to form Fabry-Perot resonators, narrow-line laser emission was also observed with the moderate threshold of ~50 kW/cm^2 at 20°K when excited with a pulsed nitrogen laser.

A study of the shift of the wavelength of the stimulated emission in magnetic fields up to 100 kG reveals that the mechanism producing this efficient emission in $CdSnP_2$:Ag is the direct radiative recombination of electrons and holes interacting in a many-body, plasma regime. The qualitative features of the stimulated emission spectrum resemble those observed in nominally pure GaAs[2,3] and InP.[4] The recombination mechanism which we have identified in $CdSnP_2$:Ag is related to the many-body screening effects,[2-4] but differs in principle from the exciton-exciton scattering[5,6] mechanism which has been proposed[7] to explain stimulated emission in these binary crystals. On account of the light electron masses in both GaAs and InP, we propose that the nature of the lasing transition in both materials can also be unambiguously identified by a study of the magnetic field dependence of the emission wavelength.

In Fig. 1 we present the photoluminescence spectra of solution-grown[8] $CdSnP_2$:Ag at 1.7°K for two values of the incident argon laser power. Whereas the very sharp, bound-exciton lines previously reported[9] near 1.004 μm were only observed at selected spots on undoped crystals, all of the features in Fig. 1 are characteristic of Ag doping and are exceptionally homogeneous over a sample's surface. At low excitation levels, there are four prominent photoluminescence bands at 1.004, 1.026, 1.072, and 1.124 μm. At high excitation levels the 1.004-μm band grows linearly in the excitation intensity and the other three bands rapidly saturate. The nature of the latter structure will be discussed in detail else-

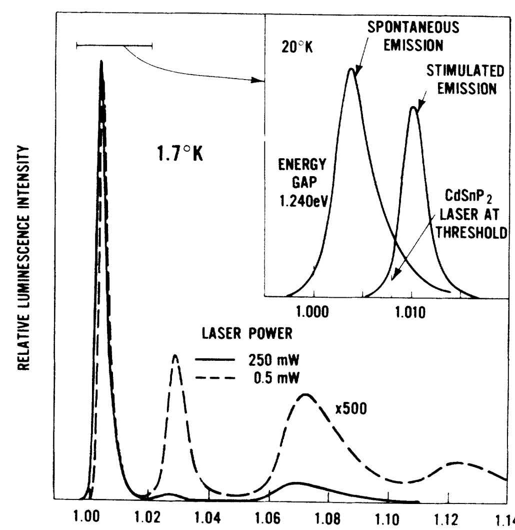

FIG. 1. Spontaneous emission spectra of Ag-doped $CdSnP_2$ for two values of the incident argon laser power. The inset compares the 1.004-μm spontaneous emission line and the stimulated spectrum excited by a pulsed nitrogen laser (~10^5 W/cm^2).

where,[10] and for the remainder of this paper we focus our attention on the 1.004-μm line which dominates at high excitation levels.

At the very high excitation levels available with a pulsed nitrogen laser, a new line, superlinearly related to pump intensity, appears in the luminescence spectra as shown in the inset to Fig. 1. Depending upon the incident intensity, the peak of the stimulated emission lies 5 to 10 meV below the spontaneous emission peak which itself lies 5 meV below the energy gap. The energy gap for $T \leq 20°$K has been located at 1.240 ± 0.002 eV by a combination of luminescence and absorption-coefficient measurements.[10]

When $CdSnP_2$:Ag crystals are cleaved to form Fabry-Perot cavities, narrow-line laser emission is observed as shown in Fig. 2. We optically pumped a cleaved $[1\bar{1}0]$ face, using the natural [112] faces to form a Fabry-Perot resonator. The experimental data in Fig. 2 were measured for pump intensities, respectively, slightly below and 15% above threshold of about 50 kW/cm^2. Single-frequency laser operation is readily obtained, indicating a homogeneously broadened gain mechanism and negligible spatial "hole burning," as we have discussed in detail for CdSe and ZnO.[11]

We identify the recombination mechanism by observing the effect of magnetic field on emission wavelength. Thus, in material with negligible carrier concentration the energy gap will increase as $\frac{1}{2}\hbar\omega_c$, where the cyclotron frequency ω_c increases linearly in the magnetic field at a rapid rate due to the light electron mass. On the other hand, the energy of a single exciton[12,13] shifts only as H^2 for small H and approaches a linear dependence at fields well above a critical field H_0 for which $\frac{1}{2}\hbar\omega_c/\mathcal{R} = 1$, where $\mathcal{R}$ is the exciton Rydberg. H_0 is about 50 kG for InP, GaAs, or $CdSnP_2$. In the exciton-exciton scattering mechanism, one exciton is annihilated and one exciton is promoted to an excited state. Hence the emitted photon is downshifted in energy by something less than one Rydberg relative to the energy of a free exciton. For fields below a few H_0, the exciton Rydberg increases linearly and since the exciton energy itself shifts only as H^2, the photon emitted as a result of exciton-exciton scattering must shift to *lower* energy. This process is thus immediately excluded for $CdSnP_2$ since observed shift with magnetic field (see Fig. 3) is to higher energy.

For our samples the carrier concentration (2.5×10^{17} cm^{-3}), electron mass ($0.06m_0$), and dielectric constant (12) are such that a weakly interacting exciton description is, in any case,

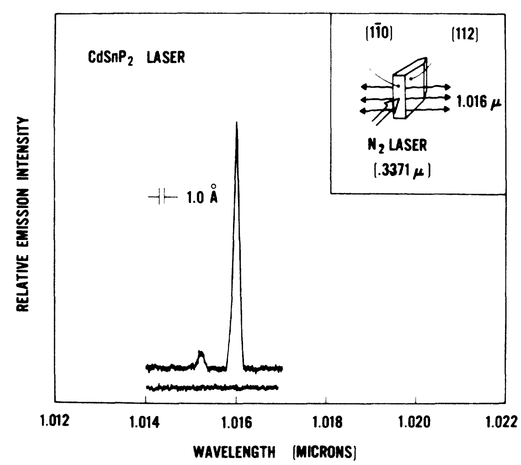

FIG. 2. Laser emission spectra at 80°K measured, respectively, slightly below and 15% above threshold of ~50 kW/cm^2. As shown in the inset, the nitrogen laser was incident onto a cleaved $[1\bar{1}0]$ face, and the natural [112] faces formed the Fabry-Perot resonator. The mode spacing agrees with cavity length (110 μm) and $n - \lambda_0\partial n/\partial\lambda = 6.3$ (Ref. 10).

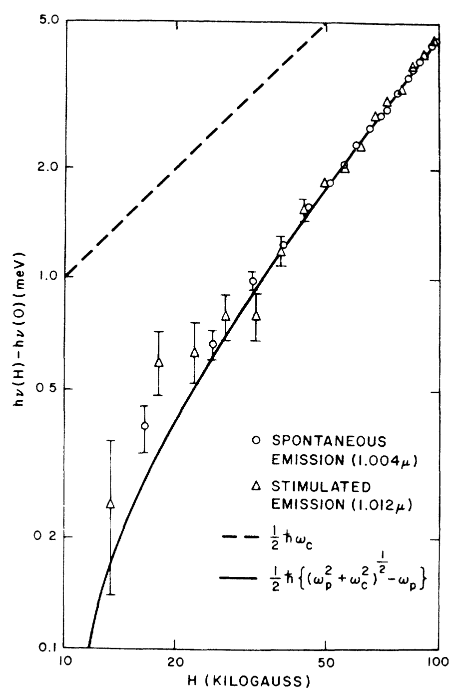

FIG. 3. Magnetic field dependence of the peaks of the spontaneous and stimulated emission spectra at 2°K. The solid line gives the theoretical shift expected for the plasmaron zero-point energy for $m^* = 0.6$, $\epsilon = 12$, and $n = 2.5\times10^{17}$ cm^{-3}. The dashed line shows the zero-point cyclotron (Landau level) shift expected for negligible carrier density.

invalid. Further, the plasma frequency and cyclotron frequency become comparable in magnitude within the range of magnetic field used experimentally. The shift of the conduction band is thus *not* given simply by the cyclotron frequency, but by the hybrid-plasmon[14] frequency (times $\hbar/2$). A detailed treatment of the theory will be published elsewhere,[15] including justification for the approximate relation we use here for the plasmon frequency[16]: $\omega^2 \sim \omega_p^2 + \omega_c^2$, with ω_p the plasma frequency. The "band-gap" shift is thus the change in $\frac{1}{2}\hbar\omega$ with magnetic field, corresponding to the shift of zero-point energy of the hybrid-plasmon field. This theoretical shift is shown, with the data, in Fig. 3. A plot of $\frac{1}{2}\hbar\omega_c$ is given for comparison. We emphasize that there are no adjustable parameters in this simple theory. An essentially identical shift is observed at 2 or 77°K.

Although space does not permit justification of our analysis here, we emphasize that the necessary conditions[15] in terms of Fermi energy, thermal energies, screening and Fermi wavelengths, carrier concentration, exciton energies and radii, etc. are indeed met under the experimental conditions, and a plasma treatment is in fact appropriate and necessary.

A (perhaps overly) simple physical picture for the shift is that each electron can give up, upon recombination, not only the zero-field energy corresponding to the band gap, but also its share of the zero-point energy of the plasmon bath. In our case essentially all electron degrees of freedom (or at least all that are of importance for radiative recombination) appear to be accounted for by the longitudinal hybrid-plasmon modes. In this connection we note that the "density parameter" r_s, defined by $4\pi r_s^3 a_0^3/3 = n^{-1}$, where n is the carrier density, is about unity for our experiments, and thus neither the high-density ($r_s \ll 1$) nor low-density ($r_s \gg 1$) treatments are strictly valid. The former theories predict that collective (i.e., plasma) modes become more important as r_s increases towards unity, and plasmons with momenta up to a bit less than half the Fermi momentum are well defined. The nature of the states between $k_F/2$ and k_F is not so clear, but our results indicate that these states are probably collective rather than single particle in nature since the latter would shift like $\hbar\omega_c/2$ in the magnetic field.[17]

The individual electron may thus be considered to interact with the other carriers collectively, i.e., via electron-plasmon, rather than electron-electron, interactions. Lundqvist[18] originally used the term "plasmaron" to refer to a resonant hole–real-plasmon state, but the term has since been generalized[16] to include all Fröhlich-type electron-plasmon interactions.[19] The radiative recombination of a "plasmaron" may thus be said to correspond to the emission process we identify in $CdSnP_2$:Ag.

We did not observe efficient stimulated emission in "nominally pure" $CdSnP_2$ which is in our case n-type with $\sim 10^{18}$ or more carriers per cm^3. There are indications that the density of sites for nonradiative recombination is much reduced in the Ag-doped material. Thus the Ag may not be acting primarily as a compensating acceptor but may also importantly affect the overall crystal quality by altering growth conditions.

In conclusion, we have observed stimulated emission and laser operation in $CdSnP_2$:Ag, which can be identified as arising from direct recombination of carriers in a many-body (plasma) regime. The magnetic field shift is quantitatively described in terms of plasmaron coupling with hybrid plasmons. This is the first evidence for a plasmaron effect in semiconductor lasers, and the first clear-cut experimental evidence for the existence of plasmaron coupling in semiconductors generally.[21] The magnetic field shift is material dependent only on m^* and ϵ, and thus the study of magnetic field shift of emission wavelength in GaAs and InP should also permit distinction between many-body and exciton-related recombination models, and provide important information concerning the physics of semiconductors at high excitation density.

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[20]Highly suggestive results were observed in InSb (Ref. 16), but the evaluation there was that the features could not be attributed to plasmaron effects "in a straightforward way."

Vortex-Free Landau State in Rotating Superfluid Helium

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Evidence is presented that below a rather large angular velocity, ω_{c1}, there is a vortex-free state in rotating locked helium, which we call the Landau state. This is similar to the Meissner state in superconductors. We further show that all the properties of the rotating helium have counterparts in irreversible type-II superconductors and find that ω_{c2} (analogous to H_{c2}) greatly exceeds our high speeds of rotation.

An important aspect of the two-fluid theory of superfluid helium as proposed by Landau[1] is that the superfluid component can flow only in an irrotational manner (i.e., $\nabla\times\vec{v}_s=0$, where $\vec{v}_s$ is the velocity of the superfluid component). The realization of stable states where $\nabla\times\vec{v}_s=0$ everywhere in the superfluid has been extremely difficult since for small velocities[2] the superfluid has a tendency to form quantized vortex lines at which $\nabla\times\vec{v}_s$ becomes singular. In this paper we report on direct observations of the stable Landau state ($\nabla\times\vec{v}_s=0$ everywhere) at rather high angular velocities for superfluid helium contained in a superleak (geometry formed by packed fine powder). It is well known that the London condition for superconductors [$\nabla\times(\vec{v}_s+q\vec{A}/mc)=0$, where $\vec{v}_s$ is the velocity of the superelectrons, $\vec{A}$ is the vector potential, and q/m is the ratio of the electron's charge to its mass] is analogous to the Landau condition for superfluid helium.[3] Thus the Meissner state of superconductors which is a consequence of the London condition is often compared with the Landau state of superfluid helium. Our experiments suggest a more precise comparison between superconductors and superfluid helium.

We have previously reported[4] on a new method of measuring the velocity of persistent currents in superfluid helium contained in a superleak. The method makes use of the fact that the speed of the wave mode (fourth sound) which propagates in clamped superfluid helium depends upon the velocity of the superfluid relative to the normal fluid (taken to be at rest) and at low temperature is given by[5]

$$C_4=C_4^{(0)}\pm(\rho_s/\rho)v_s, \tag{1}$$

where $C_4^{(0)}$ is the stationary velocity of fourth sound, ρ_s/ρ is the superfluid fraction, and v_s is the velocity of the persistent current. In Eq. (1) the plus and minus signs refer to propagation with and opposed to the persistent current. Experimentally one gets v_s from a measurement of the splitting [implied by Eq. (1)] of the resonance