

0.7 K, $\partial(C/T)\partial H \propto \text{const}H$, can be integrated to give $\chi = \chi_0[1 - 15(T/29)^2]$. Below 2 K the difference between this expression and $\chi = \chi_0[29/(T + 29)]$ is less than 2% and would not have been observed in the direct susceptibility measurements,⁵ which are dominated by the copper nuclear susceptibility below 1.3 K. The field dependence of the heat capacity, however, is very sensitive to the difference between the two expressions because it measures $\partial^2\chi/\partial T^2$, which is very different for the two expressions.

The form suggested for the temperature dependence of χ in the low-temperature single-impurity limit is particularly interesting in connection with other recent experimental results. For $T \ll T_K$, $C \propto T$,^{6,9} and recent measurements on the resistivity ρ of very dilute CuFe show¹⁴ that $\rho = \rho_0[1 - \beta(T/T_K)^2]$ where β is a constant. Thus, for CuFe, a pattern is emerging for the temperature dependences of physical properties at $T \ll T_K$ that is very different from that of a few years ago. Recent results on AuV suggest the same pattern.^{14,15} It is also interesting that several theoretical treatments of the Kondo effect predict this pattern, or parts of it.¹

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Induced Absorption in the Presence of High Electronic Excitation

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Experimental evidence is presented for the existence of an absorption continuum in CdS which is probably caused by the high density of electron-hole pairs and free excitons. The magnitude of this absorption is largest near the absorption edge and tails off toward lower energies. The presence of this continuum causes a change of the line shape of the A_1 exciton absorption.

Several semiconductors have shown stimulated light emission when excited with either photons of energy greater than the band gap or electron beam sources of high enough power. Numerous studies of this kind have been reported recently

on CdS and other II-VI materials.¹⁻⁵

Whenever the wavelength distribution of the stimulated emission as a function of the exciting light intensity is studied, one particular feature always appears to be present: At low exciting

intensity stimulated emission occurs near the band edge. With increasing exciting intensity the maximum of the stimulated emission either shifts gradually⁴ to longer wavelengths or appears sequentially at separate emission lines, always shifting to the next one because of increased⁵ excitation intensity. Temperature effects which may explain such behavior were ruled out because other observable emission lines remain equally sharp and do not shift appreciably. Apart from possibly having its cause in simple rate equations which may govern the different recombinations by stimulated emission in various centers,⁶ it also remained to be seen whether or not the absorption of the crystals remains unchanged in the corresponding wavelength region near the band edge. In other words, is the absorption constant dependent on the level of excitation? A postulated absorption change of this kind would change the observed dependence of the stimulated-emission energy on the excitation intensity. In this Letter we report our first experimental results on the change of the absorption (induced absorption) in CdS at 2°K when the crystal is excited by a high-power nitrogen laser.

The crystals were immersed in liquid He which was pumped below the λ point. The experimental arrangement is simple, in principle. The transmission of the Xe flash lamp is measured with a monochromator of 0.15-Å resolution and a 1P21 photomultiplier. At the peak of the Xe flash the nitrogen laser (337.1 nm) is triggered (pulse width of 10 nsec) and the decrease of transmitted light intensity is measured as a function of time with a sampling oscilloscope, all at constant wavelength. The induced absorption at any wavelength appeared to have two distinct time constants. The slower one has a decay time of the order of 10 μ sec for CdS platelets of about 6 μ m thickness. The magnitude of the induced absorption and its time constant varied with the crystal thickness and could be identified as caused by an increase of the lattice temperature. Samples with thicknesses between 6 and 50 μ m were investigated. The time dependence of the fast component is indistinguishable from that of the laser pulse within our resolution limits (2 nsec). The insert in Fig. 1 characterized the typical behavior. The magnitude of the fast component and its time constant did not depend on the crystal thickness.

Figure 1 shows a typical spectrum for relatively low excitation intensities. Laser power is indicated in percent of the maximal available ex-

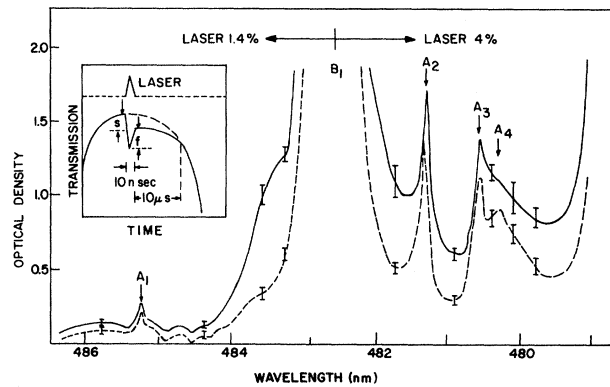


FIG. 1. The insert shows the intensity of a transmitted Xe flash. While the laser pulse is incident on the sample—schematically indicated above—the transmission shows a sudden decline of magnitude by slow (*s*) plus fast (*f*) components and a partial recovery in comparison to the transmission without simultaneous excitation by the laser (dashed line). The complete recovery of the induced absorption follows with a time constant of the order of 10 μ sec. The main figure shows the magnitude of the slow plus fast components (solid line) and of the slow component (dashed line) as functions of the wavelength in CdS platelets 6 μ m thick. The arrows mark the locations of exciton absorption in the absence of any laser excitation.

citation intensity which is 10 MW/cm². Plotted is the measured optical density as a function of the wavelength. The vertical bars indicate the maximal spread of individual measurements. A₁–A₄ and B₁ indicate the positions of the A and B exciton series absorptions in the absence of additional laser excitation.⁷ A₁ is not active in this polarization; the observed peak is due to the longitudinal component of the A exciton which is observable when the incident light is polarized with \vec{E} parallel to the crystal *c* axis. The difference between the two curves corresponds to the magnitude of the “fast” component of the induced absorption.

The center section of Fig. 2 shows the induced optical density of the fast component in the vicinity of the A₁ exciton. The upper part of Fig. 2 shows the induced absorption coefficient of the slow component. With higher and higher excitation intensity the A₁ absorption broadens and shifts toward longer wavelengths corresponding to an increased crystal temperature. A sharp rise of temperature is expected in the crystal due to the laser light absorption because its thermal capacity is very small and its thermal conductivity high at this low temperature. The process of its return to equilibrium with the bath temperature is not as fast because the

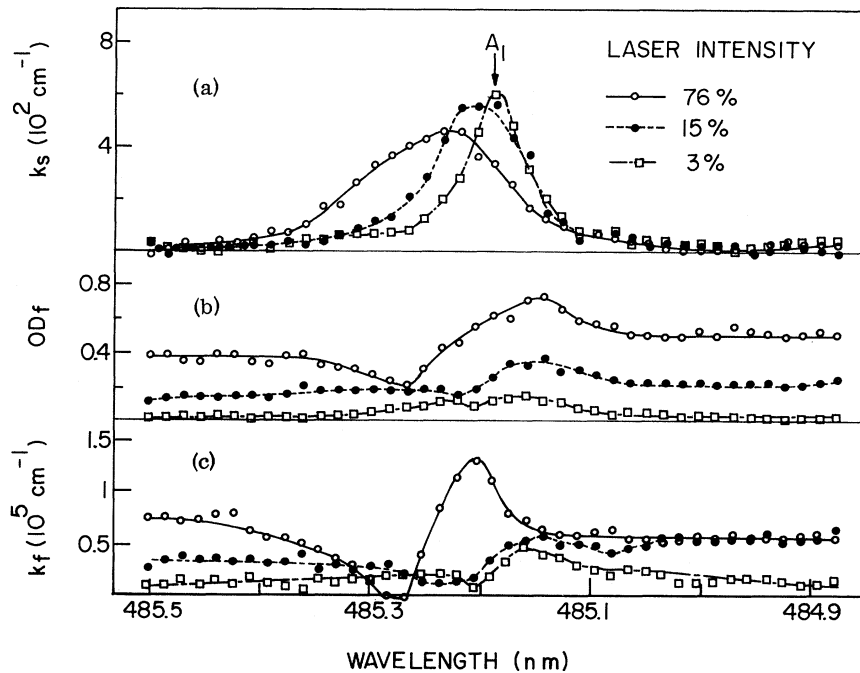


FIG. 2. (a) The induced absorption coefficient corresponding to the long-time-constant (slow-component) effect in the vicinity of the A_1 exciton; (b) the optical density associated with the fast-time-constant effect; and (c) the corresponding absorption coefficient derived with assumptions explained in the text. The CdS platelets were $20 \mu\text{m}$ thick.

main thermal resistance will occur at the CdS-He interface. Using the Kapitza resistance for NaCl we estimated that the expected time constants are in order-of-magnitude agreement with the observed ones. We therefore conclude that the induced absorption characterized by the "slow" decay is entirely due to the effect of an increase of the lattice temperature.

This, however, cannot be the case for the "fast" component of the induced absorption. We assume at the moment that it is in some way caused by the high density of electrons and holes created by the absorbed laser pulse. Without going into speculations about several possible models which could be proposed at this point, we want to emphasize here mainly two aspects of the experimental results. One is that the laser-induced "fast" absorption component increases with the excitation intensity in the spectral region near the band edge. In other words the laser excitation creates an additional background absorption which is strongest near the band edge and tails off toward longer wavelengths.

Secondly, in the immediate vicinity of absorption lines (e.g., A_1) the induced background can be considered to be wavelength independent. The shape of the absorption line, however, changes, with increasing excitation resembling one of the

curves derived by Fano⁸ for the case of interference of an autoionized state with a continuum of states. This is observable in the optical density spectrum of the fast component and even clearer at the lower part of Fig. 2, where the absorption constant related to the "fast" component is plotted. [The reduction from the optical density to the absorption constant k_f has been made under two principal assumptions: (a) All of the laser light is absorbed within 10^{-5} cm, corresponding to the absorption constant of CdS at the laser frequency; and (b) the induced absorption corresponding to the "fast" component occurs within that same dimension of the surface region.]

Hopfield, Dean, and Thomas⁹ have seen similar line shapes in N-doped GaP due to final states of an exciton bound to nitrogen and a continuum due to free excitons and electron-hole states. While in our case one could postulate, as they did, the existence of a continuum of states above the A_1 exciton due to exciton-phonon states or excitons with nonzero kinetic energy, it is not possible to extend such a process to energies below the energy of the A_1 exciton.

Thus the continuum of states induced by the high excitation density must be of a different origin. It has to fulfill the requirements, on the

other hand, that are implicit in Fano's treatment, i.e., that the absorption to these states is capable of interfering with the absorption which creates the A_1 exciton.

The main conclusion drawn from the above-described experiments is that we have observed an absorption continuum near the absorption edge which is induced by the high excitation density provided by the absorption of N-laser light. This absorption (fast component) is not due to an elevation of the crystal temperature and subsequent phonon-assisted absorption. Further, the electronic states responsible for this continuum have to be of such nature that transitions to them interfere with transitions which form the A_1 exciton.

To our knowledge, absorption due to an excitation-induced high density of electron-hole pairs and/or free excitons in intrinsic semiconductors has not been reported previously. Further experiments are in progress to gain detailed

knowledge of the intensity dependence of the induced absorption and to extend the measurements to lower energies.

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$K^\pi = 1^+$ Rotational Band in ^{18}F

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The 5298- and 6567-keV states in ^{18}F have been studied by the reaction $^{14}\text{N}(\alpha, \gamma)^{18}\text{F}$. Spin assignments have been made of $J^\pi = 4^+$ and 5^+ , respectively. The energy and spin sequence of the 1702- (1^+), 2524- (2^+), 3358- (3^+), 5298- (4^+), and 6567-keV (5^+) states, as well as enhanced $E2$ strengths of the γ transitions between these states, show that these states can be considered to be members of a predominantly four-particle, two-hole $K^\pi = 1^+$ rotational band. A comparison between existing theories is given.

There has been considerable interest recently¹⁻⁵ in the possible two-particle (2p) and four-particle two-hole (4p-2h) nature of the low-lying even-parity states in ^{18}F . Two alternative descriptions have been proposed for the 4p-2h configurations, namely, a strong-coupling¹ and weak-coupling²⁻⁴ model. Until recently neither of these two models could be supported because of the lack of reliable experimental information on the low-lying states of ^{18}F . This Letter reports experimental results which allow a clarification of the nature of the predominantly 4p-2h states.⁴

The 5298- and 6567-keV states were studied by the capture reaction $^{14}\text{N}(\alpha, \gamma)^{18}\text{F}$ at the 1135- and 2766-keV resonances, respectively. TiN targets on Ta or W backings were bombarded by a 60- to 130- μA ^4He beam from a 3-MV Van de Graaff accelerator. The γ rays were observed with 40-cm³ Ge(Li) detectors.

In addition to the known⁶ resonant γ rays from the 5298-keV level to the 937- (9%) and 2524-keV (78%) states, three new transitions to the 1123-, 3358-, and 4650-keV states were observed with respective branches of 7, 5, and 1%. The analysis of the angular distributions of these γ -ray transitions resulted in unique $J(5298) = 4$ and $J(3358) = 3$ spin assignments. The lifetime $\tau = 31 \pm 6$ fsec of the resonance state, obtained from the attenuated Doppler shift of the resonant γ rays [$F(\tau) = 0.84 \pm 0.01$] and the $\omega\gamma$ width⁷ of the resonance ($\omega\gamma = 19 \pm 7$ meV), yields $\Gamma_\gamma = \Gamma_\alpha = 12 \pm 4$ meV, in excellent agreement with the previous result⁸ of $\Gamma_\gamma/\Gamma_{\text{tot}} = 0.5 \pm 0.1$. The 78% branch to the $J^\pi = 2^+$, 2524-keV level corresponds to an $E2$ or $M2$ strength of 25 or 580 W.u. (Weisskopf units), respectively, implying $J^\pi(5298) = 4^+$.

The 6567-keV state was observed previously⁹ as a g -wave resonance, $\Gamma_\alpha \sim 900$ eV, in elastic α