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We show that even if there is no direct hole–electron competition in InP:Fe the gain is lower than expected. We attribute this reduction of the gain to a thermally induced hole–electron competition that involves the excited state of Fe^{2+} ($^5T_2$). We present a new model for the photorefractive effect in InP:Fe, which we use to interpret our beam-coupling results. Some material parameters of the InP:Fe sample and particularly of the excited state are deduced. The same energy-transfer measurements conducted in GaAs give us the ratio of hole and electron photoionization cross sections.

INTRODUCTION

In a large number of semi-insulating materials, such as sillenites, ferroelectrics, and III–V semiconductors,[1] the photorefractive effect has now been identified and studied. In general, it can be well described by a model with a single active center with one or two types of charge carrier (holes or electrons). However, in semi-insulators, for example $BaTiO_3$ and $Bi_{12}SiO_{20}$, discrepancies between theory and experimental results lead to the inclusion of secondary centers or shallow traps in models in order to increase the agreement with experiment and to be able to explain some peculiarities.[2–6] Semiconductors, in particular InP, are extremely pure compared with other materials. Thus secondary centers are not important, but, as will be shown, the Fe^{2+} excited state of iron-doped InP plays an important role in the photorefractive effect. Its introduction into the model leads to a reduction of the photorefractive gain.

We conducted cw energy transfer experiments by two-wave mixing in InP:Fe and GaAs:EL_2 crystals. For these experiments we used a diode-pumped cw Nd:YAG laser (emitting at $\lambda = 1.06\ \mu m$). The grating wave number k was directed along the (001) axis of the crystal, and the two beams were linearly polarized in the (110) direction. The results in the iron-doped InP crystal are incompatible with those predicted by the current theoretical beam-coupling model and the known parameters for our crystal. Being aware of the importance of the excited state ($^5T_2$) of Fe^{2+} in other experiments, such as nanosecond induced absorption,[7] we have introduced this excited state (called Fe^{2+*} below) into the current band-transport model. The result is a new type of hole–electron competition. Thermal emission into the conduction band empties all electrons that are excited into the Fe^{2+*} level. The effect can be called thermally induced hole–electron competition. As a consequence, we obtain a new expression for the space-charge field and the photorefractive gain. Good agreement is found when the experimental results and the gain deduced from the new expression are compared, and the determination of the concentration of Fe^{3+} is in accord with the one determined by electron paramagnetic resonance measurements. Finally, we present beam-coupling results for a sample of GaAs:EL_2 and deduce the photoionization cross section of holes and the concentrations of EL_2^0 and EL_2^+ of the crystal.

INFLUENCE OF Fe^{2+*} ON THE PHOTOREFRACTIVE EFFECT

The current model for the photorefractive effect in semiconductors takes into consideration a single active center that is coupled with both conduction and valence bands.[8,9] The resultant hole–electron competition induces a reduction of the photorefractive gain compared with that which would be obtained with a single type of carrier. We conducted two-wave mixing experiments on an InP:Fe sample grown at the Centre National d'Etudes des Télécommunications (CNET), Lannion, for which the electron contribution should be negligible. In a sample from the same boule, electron paramagnetic resonance and secondary ion mass spectroscopy measurements gave an Fe^{3+} density of $[Fe^{3+}] \simeq 8 \times 10^{16}\ cm^{-3}$ and a total iron density of $[Fe] \simeq 7 \times 10^{16}\ cm^{-3}$.[10] With the accuracy of these measurements we can estimate the upper limit of Fe^{2+} density to be $[Fe^{2+}] \leq 10^{16}\ cm^{-3}$. A similar Fe^{3+} concentration was obtained in our sample in a cross measurement.[11] The photoionization cross sections for holes (S_p) and electrons (S_n) are $S_p \simeq 5 \times 10^{-17}\ cm^2$ and $S_n \leq S_p$.[12] If we now split the total absorption into an electron (α_n) and a hole (α_p) contribution, we obtain $\alpha_n = S_n[Fe^{2+}] \ll \alpha_p = S_p[Fe^{3+}]$. This implies that the hole–electron competition is negligible. The photorefractive gain in our sample should be as high as that calculated for a single type of carrier. To our surprise, the measured photorefractive gain is much smaller than expected (Fig. 1, curves A and B).

Photoluminescence,[13] deep-level optical spectroscopy, and deep-level transient spectroscopy[14,15] have shown that Fe^{2+} exists in two states, the ^5E ground and the $^5T_2$ excited states. The latter is located 0.35 eV above the ^5E

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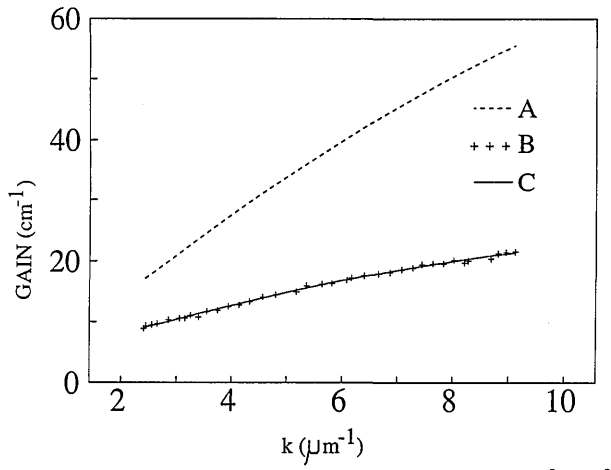

Fig. 1. Curve A, theoretical gain expected from a Fe^{2+}/Fe^{3+} level coupled with both conduction and valence bands with $\xi = 1$ ($\alpha_n \ll \alpha_p$) and $k_0^2 = 500\ \mu m^{-2}$ ($[Fe^{2+}] \simeq 10^{-16}\ cm^{-3} \ll [Fe^{3+}]$). Curve B, experimental gain. Curve C, theoretical gain with contribution of the Fe^{2+*} level considered.

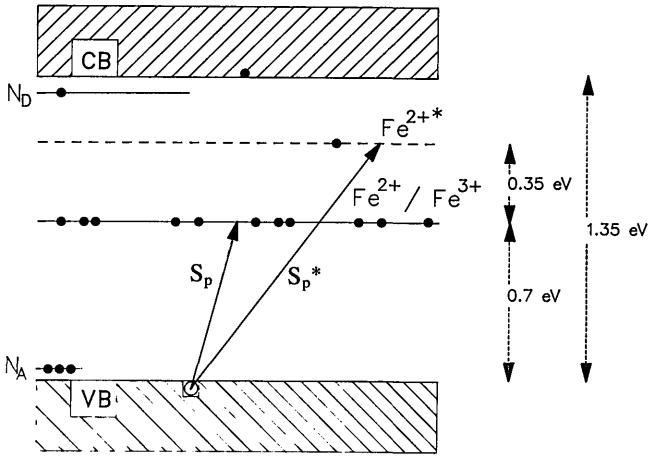

Fig. 2. Energy diagrams for InP:Fe at room temperature.

level. The energy diagram is shown in Fig. 2.[16] From this energy diagram we can deduce the influence of the Fe^{2+*} excited state on the photorefractive effect in InP:Fe. Photogeneration of holes produces both Fe^{2+} and Fe^{2+*} from Fe^{3+} with photoionization cross sections S_p and S_p^* of the same order of magnitude at the wavelength $\lambda = 1.06\ \mu m$.[7,12] The time constant of the thermal emission of the Fe^{2+*} level is quite small. It is much smaller than that of the Fe^{2+} level because of the positions of Fe^{2+} and Fe^{2+*} relative to the conduction band. Thus the electrons that are in the Fe^{2+*} level are rapidly lifted to the conduction band by thermal emission. The creation of free holes and electrons can be described as follows:

$$Fe^{2+} \xrightarrow[h\nu]{} Fe^{3+} + n_{CB},$$

$$Fe^{3+} \xrightarrow[h\nu]{} \begin{cases} Fe^{2+} + p_{VB} \\ Fe^{2+*} + p_{VB} \xrightarrow[\text{thermal}]{} Fe^{3+} + n_{CB} + p_{VB} \end{cases}.$$

So, in addition to electrons that are created directly by photoionization from Fe^{2+}, we have here a new process for the creation of free electrons. This gives rise to a new type of hole–electron competition that reduces the gain in our sample.

EXPRESSION OF THE SPACE-CHARGE FIELD

To establish the expression of the photorefractive gain, we extended Kukhtarev's model,[17] considering both photogeneration and thermal emission of electrons and holes from Fe^{3+}, Fe^{2+}, and Fe^{2+*}. In the following N_T represents the total iron density, N corresponds to $[Fe^{3+}]$, N^* to $[Fe^{2+*}]$, and $N_T - N - N^*$ to $[Fe^{2+}]$. The electron and hole current densities are $\mathbf{j}_n$, and $\mathbf{j}_p$ and the free hole and electron densities are p and n. $\mathbf{E}$ is the electric field inside the crystal. The material equations are

the rate equations for $[Fe^{3+}]$ and $[Fe^{2+*}]$,

$$\begin{aligned} \partial N/\partial t &= (\beta_n + S_n I)(N_T - N - N^*) - \gamma_n n N \\ &\quad + (\beta_n^* + S_n^* I)N^* - \gamma_n^* n N - (\beta_p + S_p I)N \\ &\quad + \gamma_p p(N_T - N - N^*) \\ &\quad - (\beta_p^* + S_p^* I)N + \gamma_p^* p N^*, \\ \partial N^*/\partial t &= -(\beta_n^* + S_n^* I)N^* + \gamma_n^* n N \\ &\quad + (\beta_p^* + S_p^* I)N - \gamma_p^* p N^*; \end{aligned}$$

the current density equations,

$$\mathbf{j}_n = e n \mu_n \mathbf{E} + \mu_n k_B T\ \mathbf{grad}\ n,$$

$$\mathbf{j}_p = e p \mu_p \mathbf{E} - \mu_p k_B T\ \mathbf{grad}\ p;$$

the continuity equations,

$$\begin{aligned} \partial n/\partial t &= \operatorname{div}(\mathbf{j}_n/e) + (\beta_n + S_n I)(N_T - N - N^*) - \gamma_n n N \\ &\quad + (\beta_n^* + S_n^* I)N^* - \gamma_n^* n N, \\ \partial p/\partial t &= -\operatorname{div}(\mathbf{j}_p/e) + (\beta_p + S_p I)N - \gamma_p p(N_T - N - N^*) \\ &\quad + (\beta_p^* + S_p^* I)N - \gamma_p^* p N^*; \end{aligned}$$

and the Poisson equation

$$\operatorname{div}\mathbf{E} = (-e/\varepsilon)(N_T - N - N_D + N_A + n - p).$$

I is a spatially modulated illumination (photon current in $sec^{-1}\ cm^{-2}$), $I(z) = I_0\{1 + \mathrm{Re}[m \exp(ikz)]\}$, e, μ_n, and μ_p are the absolute values of electron and hole charges and mobilities; ε is the static dielectric constant of the material ($\varepsilon = \varepsilon_0 \varepsilon_R$ with[18] $\varepsilon_R = 12.7$), and N_D and N_A are the density of compensative donors and acceptors, respectively. In these equations S represents the photoionization cross sections, γ the recombination coefficients, and β the thermal emission rates. The indices n, p, and * stand for electron, hole, and Fe^{2+*}, respectively.

Considering small modulations of the illumination pattern $m \ll 1$, we perform the usual linearization of the previous equations, keeping only the zeroth-order term and the spatially modulated first-order term, expressed as

$$X(t) = X_0 + \mathrm{Re}[X_1(t)\exp(ikz)].$$

A general analytical solution can be found, for quasi-cw illumination. The dark conductivity is due mainly to electrons.[10,16] The thermal emission rate[19] β_n is $\sim 10\ sec^{-1}$ at ambient temperature (300 K). Since thermal emission rates depend exponentially on the energy difference between the levels and the conduction band, β_n^* ($\simeq 10^7\ sec^{-1}$) is 6 orders of magnitude larger than β_n. This is an approximate value, since it was calculated with the same parameters as β_n except level energy. The rate β_p is com-

puted from $p = [Fe^{3+}]\beta_p\tau_p$ with $p < 10^7$ cm^{-3} (electrons are dominant in dark current) and the recombination time of holes on Fe^{2+}, $\tau_p \simeq 120$ nsec,[20] giving $\beta_p \simeq 10^{-4}$ sec$^{-1} \ll \beta_n$. For an illumination of several tens of milliwatts, the dark current in the conduction band is smaller than or approximately equal to the electron photocurrent; thus $\beta_n \leq S_nI_0$; β_p, $\beta_p^* \ll S_pI_0$, $S_p^*I_0$, and $\beta_n^* \gg S_n^*I_0$. Assuming equal recombination coefficients ($\gamma_p \simeq \gamma_p^*$) for holes on Fe^{2+} and Fe^{2+*}, one has $\beta_n^* \gg \gamma_p^*p_0$ ($\simeq 10^2$ sec^{-1} for $I_0 \times h\nu \simeq 100$ mW cm^{-2}). This means that Fe^{2+*} is changed into Fe^{3+} primarily by thermal excitation of electrons and not by hole recombination. For low irradiance the population of the Fe^{2+*} level is always extremely small, $N_0^* \ll N_0, N_T - N_0$. We now make the usual simplifications for low irradiance:

$$n_0, p_0 \ll N_0, N_\tau - N_0,$$
$$n_1, p_1 \ll N_1.$$

Moreover, the times considered in quasi-cw illumination are much longer than the lifetimes of Fe^{2+*} and both carriers; so we have

$$\frac{\partial n_1}{\partial t} = \frac{\partial p_1}{\partial t} = \frac{\partial N_1^*}{\partial t} = 0.$$

The resolution of the equations yields a first-order differential equation that governs the spatially modulated part of the field E_1:

$$-\varepsilon\left(\frac{k_BT}{e^2I_0}\right)\frac{\partial E_1}{\partial t} = E_1\left\{A_n\alpha_n\frac{[k(k - iV)/k_0^2] + 1}{k(k - iV) + \kappa_n^2} + (\alpha_p + \alpha_p^*)\frac{[k(k + iV)/k_0^2] + 1}{k(k + iV) + \kappa_p^2} + \frac{\alpha_p^*}{k(k - iV) + \kappa_n^2}\right\} + \mathrm{Im}\left(\frac{k_BT}{e}\right)\left[(\alpha_n + \alpha_p^*)\frac{k - iV}{k(k - iV) + \kappa_n^2} - (\alpha_p + \alpha_p^*)\frac{k + iV}{k(k + iV) + \kappa_p^2}\right], \quad (1)$$

with

$$k_0^2 = \frac{e^2}{\varepsilon k_BT}\frac{N_0(N_T - N_0)}{N_T},$$

$$\kappa_n^2 = \frac{e}{k_BT}\frac{\gamma_nN_0}{\mu_n} = \frac{e}{k_BT}\frac{1}{\mu_n\tau_n},$$

$$\kappa_p^2 = \frac{e}{k_BT}\frac{\gamma_p(N_T - N_0)}{\mu_p} = \frac{e}{k_BT}\frac{1}{\mu_p\tau_p},$$

$$\alpha_n = S_n(N_T - N_0), \quad \alpha_p = S_pN_0, \quad \alpha_p^* = S_p^*N_0,$$
$$A_n = (\beta_n + S_nI_0)/S_nI_o, \quad V = eE_0/k_BT,$$

where E_0 is the external applied field and κ_n and κ_p are the inverse diffusion lengths for electrons and holes, respectively.

The quasi-cw concentrations of free electrons, free holes, Fe^{3+}, and Fe^{2+*} are

$$n_0 = \frac{(\beta_n + S_nI_0)(N_T - N_0) + S_p^*I_0N_0}{\gamma_nN_0},$$

$$p_0 = \frac{(S_p^* + S_p)I_0N_0}{\gamma_p(N_T - N_0)}, \quad N_0 = N_T - (N_D - N_A),$$

$$N_0^* = \frac{\gamma_n^*}{\gamma_n\beta_n^*}(\beta_n + S_nI_0)(N_T - N_0) + \frac{(\gamma_n + \gamma_n^*)}{\gamma_n\beta_n^*}S_p^*I_0N_0.$$

Every occupied Fe^{2+*} state almost immediately generates an electron by thermal excitation. Thus the influence of the Fe^{2+*} state in the differential equation for E_1 is visible only by the term α_p^*, the absorption that gives rise to the photoexcitation of holes with the final level Fe^{2+*}.

For steady state, we can derive the following expression for the space-charge field, taking $\kappa_n^2, \kappa_p^2 \ll k_0^2$ (which is obvious in semiconductors):

$$E_1 = \mathrm{Im}\left(\frac{k_BT}{e}\right) \times \frac{k(k^2 + V^2)(\alpha_p - \alpha_n) + k[(\alpha_p + \alpha_p^*)\kappa_n^2 - (\alpha_n + \alpha_p^*)\kappa_p^2] + iV[(\alpha_p + \alpha_p^*)\kappa_n^2 + (\alpha_n + \alpha_p^*)\kappa_p^2]}{[(A_n\alpha_n + \alpha_p^*)\kappa_p^2 + (\alpha_p + \alpha_p^*)\kappa_n^2] + k^2(A_n\alpha_n + \alpha_p + 2\alpha_p^*) + [k^2(k^2 + V^2)/k_0^2](A_n\alpha_n + \alpha_p + \alpha_p^*) + ikV[A_n\alpha_n - \alpha_p]}. \quad (2)$$

This is the complex expression of the space-charge field in the presence of an external field. For small grating spacings (i.e., large grating wave number k) we can neglect the influence of the diffusion lengths ($\kappa_n, \kappa_p \ll k$). Then the expression for the space-charge field becomes

$$E_1 = \mathrm{Im}\left(\frac{k_BT}{e}\right)\frac{k(k^2 + V^2)(\alpha_p - \alpha_n)}{k^2(A_n\alpha_n + \alpha_p + 2\alpha_p^*) + [k^2(k^2 + V^2)/k_0^2](A_n\alpha_n + \alpha_p + \alpha_p^*) + ikV(A_n\alpha_n - \alpha_p)}. \quad (3)$$

The temperature-dependent resonance of the photorefractive gain in the presence of an external field that was observed and described by Picoli *et al.*[21] also appears in this expression. For $A_n\alpha_n - \alpha_p = 0$, the space-charge field becomes purely imaginary and increases proportionally to $(k^2 + V^2)$ and thus with the external field. The condition corresponds to $(\beta_n + S_nI_0)[Fe^{2+}] = S_p[Fe^{3+}]I_0$ or, with $\beta_n \gg S_nI_0$, to $\beta_n[Fe^{2+}] = S_pI_0[Fe^{3+}]$. This is the expression found in Ref. 21, with the only difference being that here S_p is not the total hole photoionization cross section ($S_p + S_p^*$) but is only the part responsible for the ionization of holes with the final level Fe^{2+}. The condition for resonance is slightly different from that predicted by Picoli *et al.*, but the behavior of the resonance with respect to the illumination and the temperature is still the same.

Without an applied field, the steady-state space-charge field E_1 is always purely imaginary and can be written as $E_1 = \mathrm{Im}\, E_{sc}$. This 90° phase-shifted space-charge field creates an energy transfer by the electro-optic effect.

The gain is[17]

$$\Gamma = 2\pi \frac{n^3 r_{41}}{\lambda \cos\theta} E_{sc}, \tag{4}$$

where n is the refractive index, r_{41} the electro-optic coefficient, and θ the half-angle between the two beams inside the crystal.

From Eqs. (2) and (4) we derive

$$\Gamma = \frac{-2\pi n^3 r_{41}}{\lambda \cos\theta}\left(\frac{k_B T}{e}\right)\left\{k\left[\left(\frac{\alpha_n - \alpha_p}{\alpha_T + \alpha_p^*}\right)k^2 + \frac{(\alpha_n + \alpha_p^*)\kappa_p^2 - (\alpha_p + \alpha_p^*)\kappa_n^2}{(\alpha_T + \alpha_p^*)}\right]\Big/\left[k^2 + \frac{k^4}{k_0^2}\left(\frac{\alpha_T}{\alpha_T + \alpha_p^*}\right) + \frac{(\alpha_n + \alpha_p^*)\kappa_p^2 + (\alpha_p + \alpha_p^*)\kappa_n^2}{(\alpha_T + \alpha_p^*)}\right]\right\}. \tag{5}$$

We took $A_n = 1$. This approximation is justified because the measured gain does not increase with increasing intensity of the illumination, or, in other words, because the gain is saturated with respect to intensity. The symbol α_T denotes the total absorption of the sample; $\alpha_T = \alpha_n + \alpha_p + \alpha_p^*$.

BEAM COUPLING IN InP

We measured the photorefractive beam coupling gain Γ as a function of the grating spacing. The grating spacing was varied from 0.7 to 3 μm. Within this range the grating wave number k is always larger than or approximately equal to the inverse of the electron or hole diffusion lengths, $k \geq \kappa_n, \kappa_p$. For the spacings where $k^2 \gg \kappa_n^2, \kappa_p^2$, Eq. (5) can be simplified to

$$|\Gamma| = |A|\frac{\xi_0 k}{1 + (k^2/k_0'^2)},$$

with

$$A = 2\pi \frac{n^3 r_{41}}{\lambda \cos\theta}\left(\frac{k_B T}{e}\right),$$

$$\xi_0 = \frac{\alpha_n - \alpha_p}{\alpha_T + \alpha_p^*},$$

$$k_0'^2 = k_0^2\left(\frac{\alpha_T + \alpha_p^*}{\alpha_T}\right).$$

By plotting our experimental results in the form Ak/Γ versus k^2, we obtain a straight line for the large values of k^2 (Fig. 3). A is a known quantity, calculated with the values of $n, r_{41}, \lambda, k_B T/e$, and $\cos\theta$. The intercept at the origin and the slope of the straight line give $1/\xi_0$ and $1/k_0'^2\xi_0$, respectively.

For small values of k, the previous approximation is no longer valid. Indeed, for these k values the curve departs from the straight line. In this case Eq. (5) cannot be simplified. We have fitted our results with the following function for all grating spacings:

$$|\Gamma| = |A|k\frac{|\xi_0 k^2 + b - c|}{k^2 + (k^4/k_0'^2) + b + c},$$

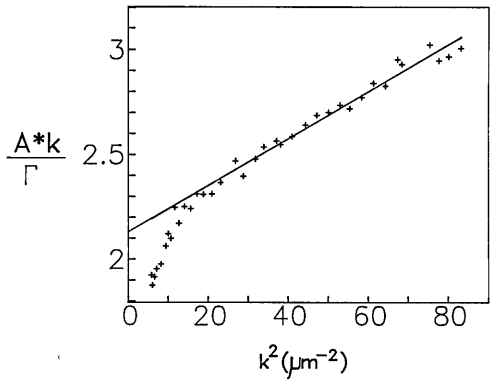

Fig. 3. Measured beam-coupling gain Γ (+) plotted in the form Ak/Γ as a function of k^2 for InP:Fe with a fitted straight line. $A = 7.11 \times 10^{-2}$ for Γ in inverse centimeters and for k in inverse micrometers, $n_0 = 3.29$, and $r_{41} = 1.34$ pm V^{-1}.[18]

with

$$b = \left(\frac{\alpha_n + \alpha_p^*}{\alpha_T + \alpha_p^*}\right)\kappa_p^2, \qquad c = \left(\frac{\alpha_p + \alpha_p^*}{\alpha_T + \alpha_p^*}\right)\kappa_n^2,$$

and $|A|$, ξ_0, and $k_0'^2$ as defined above. By iteration, one finds the parameters giving the best fit, as shown in Fig. 1, curves B and C:

$$\xi_0 = -0.424 \pm 0.03,$$

$$k_0'^2 = 260 \pm 50\ \mu\text{m}^{-2},$$

$$b \leq 0.05\ \mu m^{-2},$$

$$c = 1.4 \pm 0.5\ \mu\text{m}^{-2}.$$

The sign of ξ_0 was determined by an additional experiment, as described in Refs. 22 and 23. The data can be perfectly fitted with three parameters, since b can be set to zero without a visible effect. By including b in the fitting process, we get only an upper limit for its value.

We note that the values we found here for $k_0'^2$ and ξ_0 are close to those determined by Valley *et al.* in their beam-coupling experiments on another InP:Fe crystal.[24]

With the above set of parameters and the value of the total absorption $\alpha_T = 1.95 \pm 0.04$ cm^{-1} that we measured separately, we can now determine α_p, α_p^*, k_0^2, κ_n^2, and κ_p^2. Absorption α_n has been neglected, since in our crystal we have $\alpha_n \ll \alpha_p, \alpha_p^*$, as previously discussed. We obtain

$$\alpha_p = 1.16 \pm 0.05\ \text{cm}^{-1},$$

$$\alpha_p^* = 0.79 \pm 0.05\ \text{cm}^{-1},$$

$$k_0^2 = 185 \pm 40\ \mu\text{m}^{-2},$$

$$\kappa_n^2 = 2 \pm 1.5\ \mu\text{m}^{-2},$$

$$\kappa_p^2 \leq 0.2\ \mu\text{m}^{-2}.$$

The values of α_p and α_p^* can be expressed as percentages of the total hole absorption $\alpha_p^{tot} = \alpha_p + \alpha_p^*$:

$$\alpha_p/\alpha_p^{tot} = S_p/S_p^{tot} = (60 + 3)\%,$$

$$\alpha_p^*/\alpha_p^{tot} = S_p^*/S_p^{tot} = (40 + 3)\%.$$

S_p^{tot} corresponds to the total photoionization cross section ($S_p^{tot} = S_p + S_p^*$). This parameter was measured by

deep-level optical spectroscopy as $S_p^{tot} = 5 \times 10^{-17}$ cm^2.[12] With this value we calculate the Fe^{3+} concentration and obtain $[Fe^{3+}] = 4 \times 10^{16}$ cm^{-3}. This value is slightly lower than the value given by CNET but is in perfect accord with a study on the induced nanosecond absorption done on this same crystal.[7]

From k_0^2 we calculate the effective trap density

$$N_{\text{eff}} = [Fe^{2+}][Fe^{3+}]/([Fe^{2+}] + [Fe^{3+}]),$$

which is equal to $[Fe^{2+}] = (3.2 \pm 0.5) \times 10^{15}$ cm^{-3}.

The results for κ_n^2 and κ_p^2 give only estimates of the diffusion lengths. However, their values are in good accordance with those calculated from values of the mobilities found in the literature[10] and the recombination times for electrons and holes on iron in InP.[20]

BEAM COUPLING IN GaAs

We performed the same beam-coupling experiment as above in an undoped GaAs:EL_2 sample. The experimental result can be well explained by a model with one deep level and with electrons and holes as charge carriers. The theoretical expression for the gain is deduced from Eq. (5), with $\alpha_p^* = 0$ (no excited state in GaAs):

$$\Gamma = -2\pi \frac{n^3 r_{41}}{\lambda \cos\theta}\left(\frac{k_B T}{e}\right)\left\{k\left[\left(\frac{\alpha_n - \alpha_p}{\alpha_n + \alpha_p}\right)k^2 + \left(\frac{\alpha_n \kappa_p^2 - \alpha_p \kappa_n^2}{\alpha_n + \alpha_p}\right)\right] \Big/ \left[k^2 + \frac{k^4}{k_0^2} + \frac{\alpha_n \kappa_p^2 + \alpha_p \kappa_n^2}{\alpha_n + \alpha_p}\right]\right\}.$$

We use the same technique as that used for InP to analyze our experimental results. For GaAs the experimental curve $Ak/\Gamma = f(k^2)$ (Fig. 4) is a straight line down to 10 μm^{-2}. The small departure at even lower k values does not permit the determination of κ_n^2 and κ_p^2. From the parameters of the straight line, we obtain $|\xi_0| = 0.74 \pm 0.05$ and $k_0'^2 = 91 \pm 10$ μm^{-2}. The measured absorption of the crystal at $\lambda = 1.06$ μm is $\alpha = 0.95 \pm 0.05$ cm^{-1}. We consider, as is generally accepted, that electrons are the dominant carriers for the photorefractive effect in GaAs:EL_2. Therefore ξ_0 is positive. Then we deduce hole and electron absorptions α_p and α_n, and

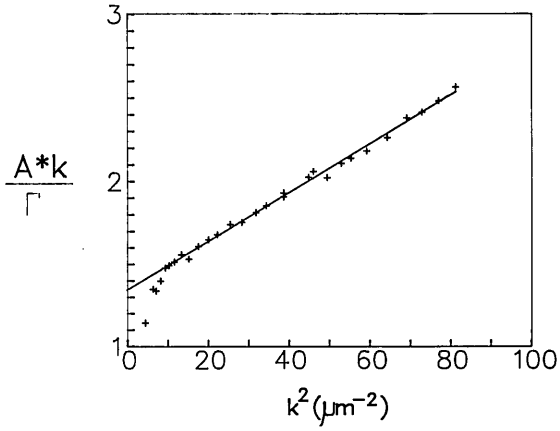

Fig. 4. Measured beam-coupling gain Γ (+) plotted in the form Ak/Γ as a function of k^2 for GaAs with a fitted straight line. $A = 8.34 \times 10^{-2}$ for Γ in inverse centimeters and k in inverse micrometers, $n_0 = 3.48$, and $r_{41} = 1.43$ pm V^{-1}.[25]

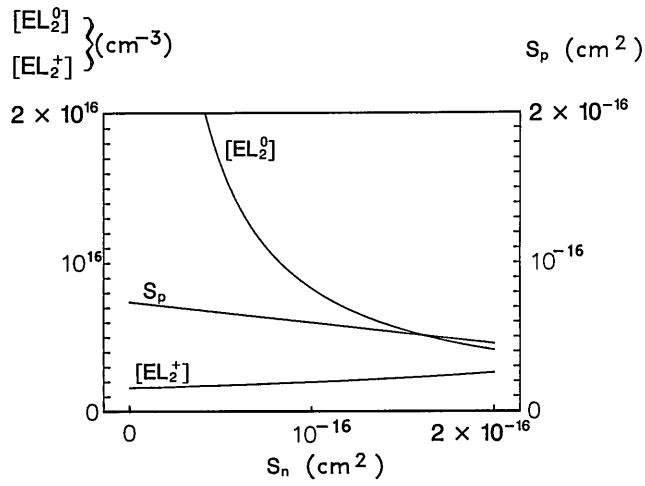

Fig. 5. Variation of S_p, $[EL_2^0]$, $[EL_2^+]$ as a function of S_n.

the effective trap density from $k_0'^2 = k_0^2$ (with $\varepsilon = \varepsilon_0 \varepsilon_R$ and $\varepsilon_R = 12.9$) (Ref. 25):

$$\alpha_n = S_n[EL_2^0] = 0.83 \pm 0.05 \text{ cm}^{-1},$$

$$\alpha_p = S_p[EL_2^+] = 0.12 \pm 0.01 \text{ cm}^{-1},$$

$$N_{\text{eff}} = \frac{[EL_2^+][EL_2^0]}{[EL_2^+] + [EL_2^0]} = (1.6 \pm 0.2) \times 10^{15} \text{ cm}^{-3}.$$

We have three relations among the four parameters S_n, S_p, $[EL_2^0]$, and $[EL_2^+]$, and we can plot three of them as a function of S_n (Fig. 5). As soon as one of the four parameters is fixed, all four are known. The plot shows the weak dependence of S_p and $[EL_2^+]$ on the value of S_n; so we can consider that this method gives a good order of magnitude for S_p and $[EL_2^+]$. Taking the well-accepted value $S_n = 1 \times 10^{-16}$ cm^2 from Ref. 26, we obtain the following set of parameters: $S_p = 6 \times 10^{-17}$ cm^2, $[EL_2^0] = 8 \times 10^{15}$ cm^{-3}, and $[EL_2^+] = 2 \times 10^{15}$ cm^{-3}.

The same experiments were reported in Ref. 27 for different samples of one ingot. The concentration of $[EL_2^0]$ and $[EL_2^+]$ and their ratio varied for the different samples, leading to different values of ξ_0 (i.e., different strengths of hole–electron competition). The above analysis can be applied to the two samples that show hole–electron competition (seed center and seed side). Taking $r_{41} = 1.43$ pm V^{-1} and $S_n = 1 \times 10^{-16}$ cm^2, we can deduce that for these two samples values of S_p are close to the one found for our sample: $S_p = 5.5 \times 10^{-17}$ cm^2 for seed center and $S_p = 5.75 \times 10^{-17}$ cm^2 for seed side. Moreover, the ratio $S_p/S_n = 0.6$ that was determined for our crystal is similar to the one found by Valley *et al.*[26] with their picosecond technique ($S_p/S_n = 0.76$).

CONCLUSION

This study confirms the importance of the excited state of Fe^{2+} for the photorefractive effect.[28] The excited state induces a drastic limitation of the photorefractive gain through a thermal hole–electron competition. This limitation will disappear when the photon energy is no longer sufficient to raise electrons from the valence band to the

Fe^{2+}* level; i.e., $h\nu < 1.05$ eV. For these wavelengths the model without the excited state will correctly describe the photorefractive effect.[29] Another possibility for suppressing this hole–electron competition is a lowered temperature. If the inverse of the thermal emission rate $[\beta_n^* = KT^2\exp(-E/k_BT)$ (Ref. 19)] is larger than the recombination time of holes on Fe^{2+}*, the Fe^{2+}* level will no longer be emptied by thermal emission of electrons in the conduction band but by recombination of holes from the valence band. Then Fe^{2+}* will play exactly the same role as Fe^{2+}, and the thermally induced hole–electron competition will disappear. The transition from one behavior to the other should occur near 170 K. A temperature-dependent study of the photorefractive effect would provide more information.

The study of undoped GaAs permitted us to determine by a cw photorefractive experiment the photoionization cross section of holes, which is not well known. A comparison with another experiment of the same type for other GaAs crystals confirms our findings. We find a ratio $S_p/S_n = 0.6$, which is close to the value found by a picosecond pump–probe technique.

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