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Observation of Intraband Coherence in the Nonlinear Optical Response of Gallium Arsenide

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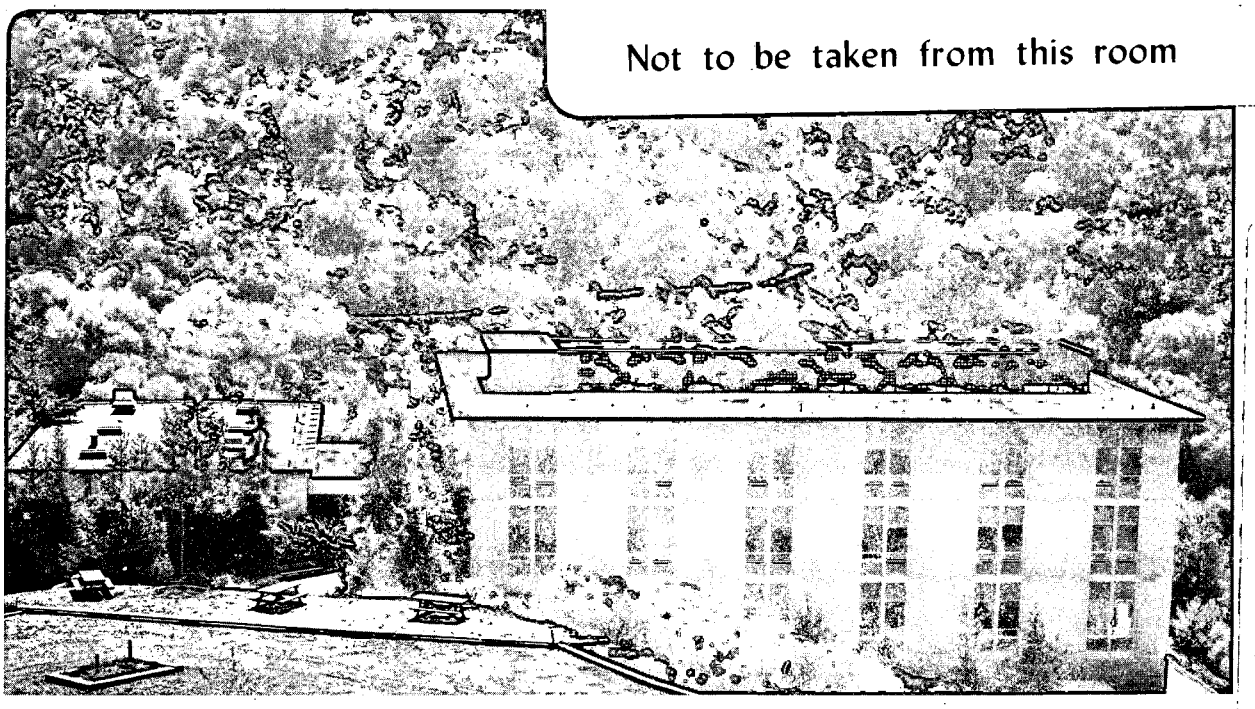

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ABSTRACT

We show that magnetic-field-induced modulation of the intraband density matrix elements produces new resonances in the power spectra of ultrashort-pulse four-wave mixing in semiconductors.

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The top of Fig. 1 presents the linear absorption spectra of a high-quality GaAs sample at $B = 8\,\mathrm{T}$ and $T = 1.6\,\mathrm{K}$. Stress has lifted the degeneracy between the light-hole (lh) and the heavy-hole (hh) subbands. Lorentzian exciton resonances are seen at the onset of the lowest (n=1) Landau transition and asymmetric Fano resonances at the higher (n>1) ones [1]. The bottom of Fig. 1 shows the spectrally resolved ultrashort pulse σ^-/σ^- Four-Wave-Mixing (FWM) signal obtained at the same field for $\Delta t = -50\,\mathrm{fs}$ and $-530\,\mathrm{fs}$ for an excitation resonant with the n=1 excitons. Strong contributions of the lh and hh excitons are clearly seen, as well as smaller ones from the n=2 Fano resonance. The FWM power spectra, however, exhibit other strong peaks at energies where no special feature is observed in the absorption spectrum. The energy of these additional peaks depends on the magnetic field, as shown in Fig. 2, where the Δt=0 fs spectra for $B = 0$, 4, 8, and 10 T are presented. No usual one- or two-photon process can explain these features.

We attribute the new resonances to a magnetic-field-induced modulation of the *intraband* density matrix elements. The density matrix $n_{ij\,s_1 s_2}(\mathbf{r}_1, \mathbf{r}_2, t)$ (i,j = c v) is a two-point function [2]. A quantum mechanical coherence between point $\mathbf{r}_1$ and point $\mathbf{r}_2$ is called an "off-diagonal long range order" [3,4]. It exists whenever $n_{jj}(\mathbf{r}_1, \mathbf{r}_2) \neq 0$. Similarly a "spin coherence" exists whenever $n_{jj\,s_1 s_2} \neq 0$. The time development of the first-order polarization is governed by the dipole excitation $-\mu\, E(t)\, \delta(\mathbf{r}_1 - \mathbf{r}_2)$, and does not involve *intraband* terms. Therefore *intraband* coherence cannot be observed in linear optics. On the contrary, such a coherence can be seen in nonlinear optics. For example the source term of the third-order polarization responsible for FWM is given by:

$$-\mu\, E(t)\,[\,n^{(2)}_{\mathrm{vv}\,s_1 s_2}(\mathbf{r}_1, \mathbf{r}_2, t) - n^{(2)}_{\mathrm{cc}\,s_1 s_2}(\mathbf{r}_1, \mathbf{r}_2, t)\,] - \sum_s \int d^3\mathbf{r}\, \Big[V(|\mathbf{r}_1 - \mathbf{r}|) - V(|\mathbf{r} - \mathbf{r}_2|)\Big] \times$$

$$\times \Big[n^{(2)}_{\mathrm{cc}\,s_1 s}(\mathbf{r}_1, \mathbf{r}, t)\, n^{(1)}_{\mathrm{cv}\,s s_2}(\mathbf{r}, \mathbf{r}_2, t) + n^{(1)}_{\mathrm{cv}\,s_1 s}(\mathbf{r}_1, \mathbf{r}, t)\, n^{(2)}_{\mathrm{vv}\,s s_2}(\mathbf{r}, \mathbf{r}_2, t)\Big]\,.$$

Here $V(r)$ is the Coulomb potential which provides a coupling between *intraband* and *interband* terms. This qualitative argument can be rigorously derived in the framework of the semiconductor Bloch equations [5].

In an unperturbed semiconductor at equilibrium, there are no specific features that reveal the coherence of n_{cc} and n_{vv}. In a magnetic field, however, they are modulated at cyclotron

frequencies or at spin-flip frequencies. This results in new resonance features observable in nonlinear optical processes. Such modulation at a cyclotron frequency has been observed in THz emission under magnetic field by Some and Nurmikko [6].

In conclusion, we report the observation of magnetic-field induced intraband coherence observed through third order nonlinear optical effects.

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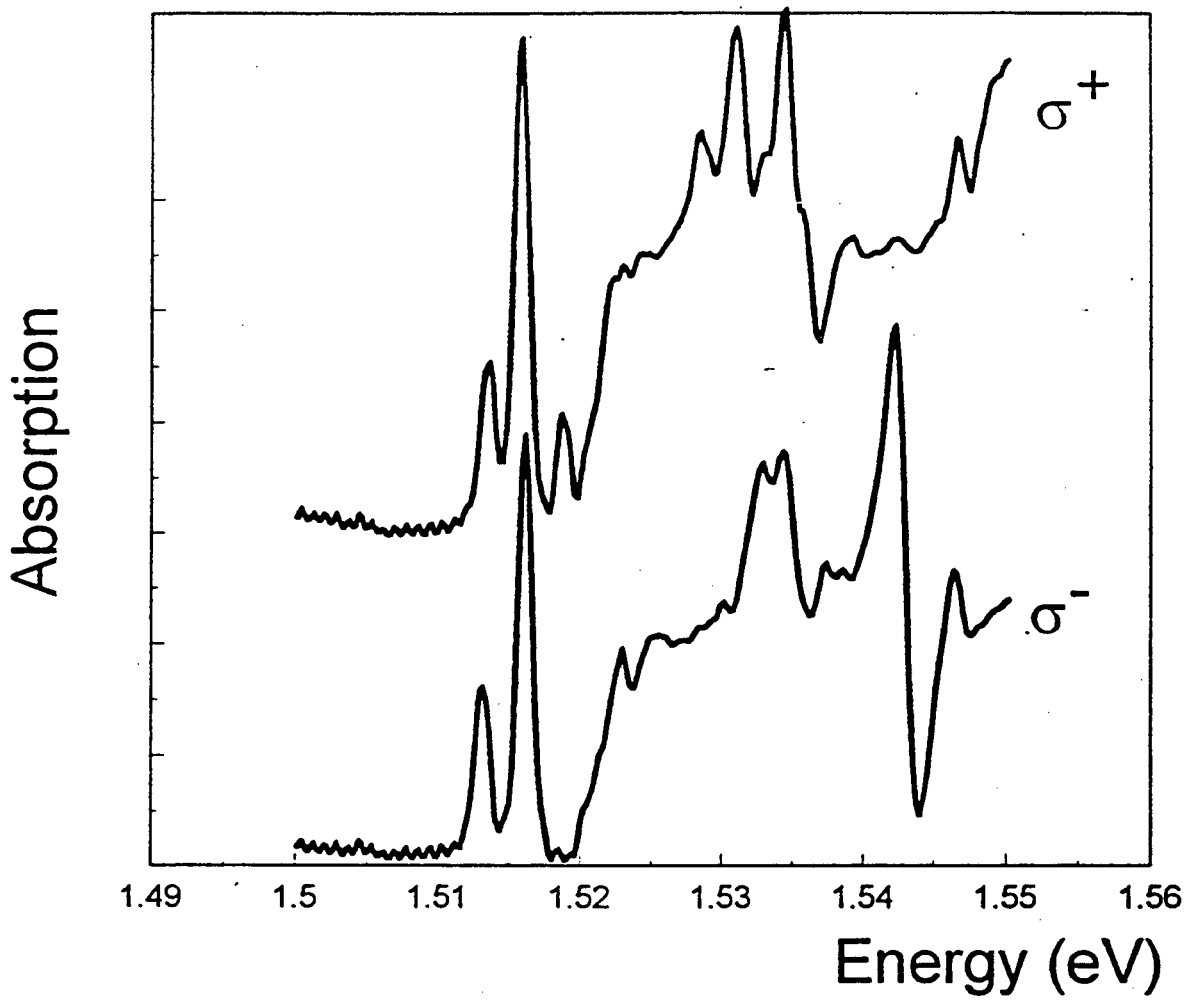
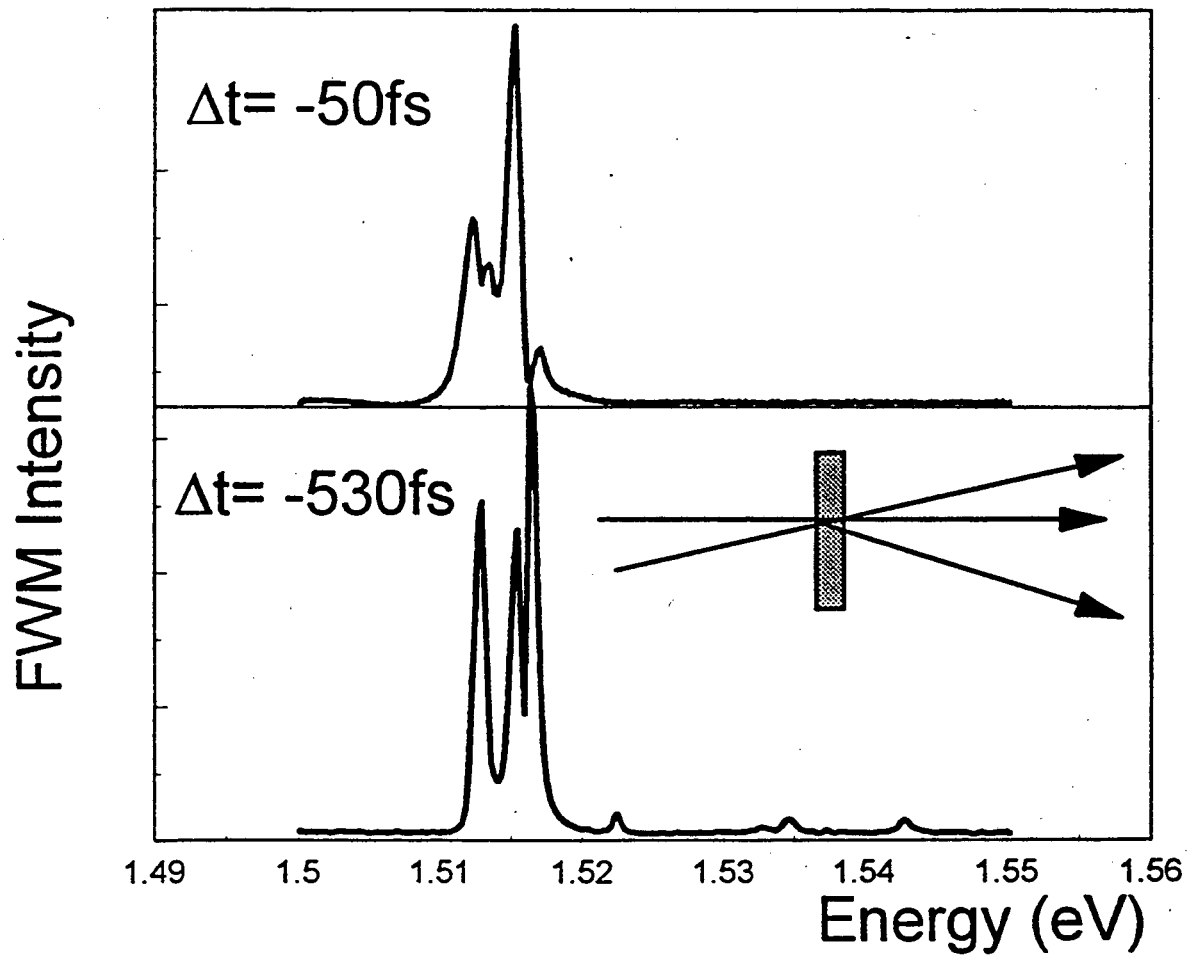
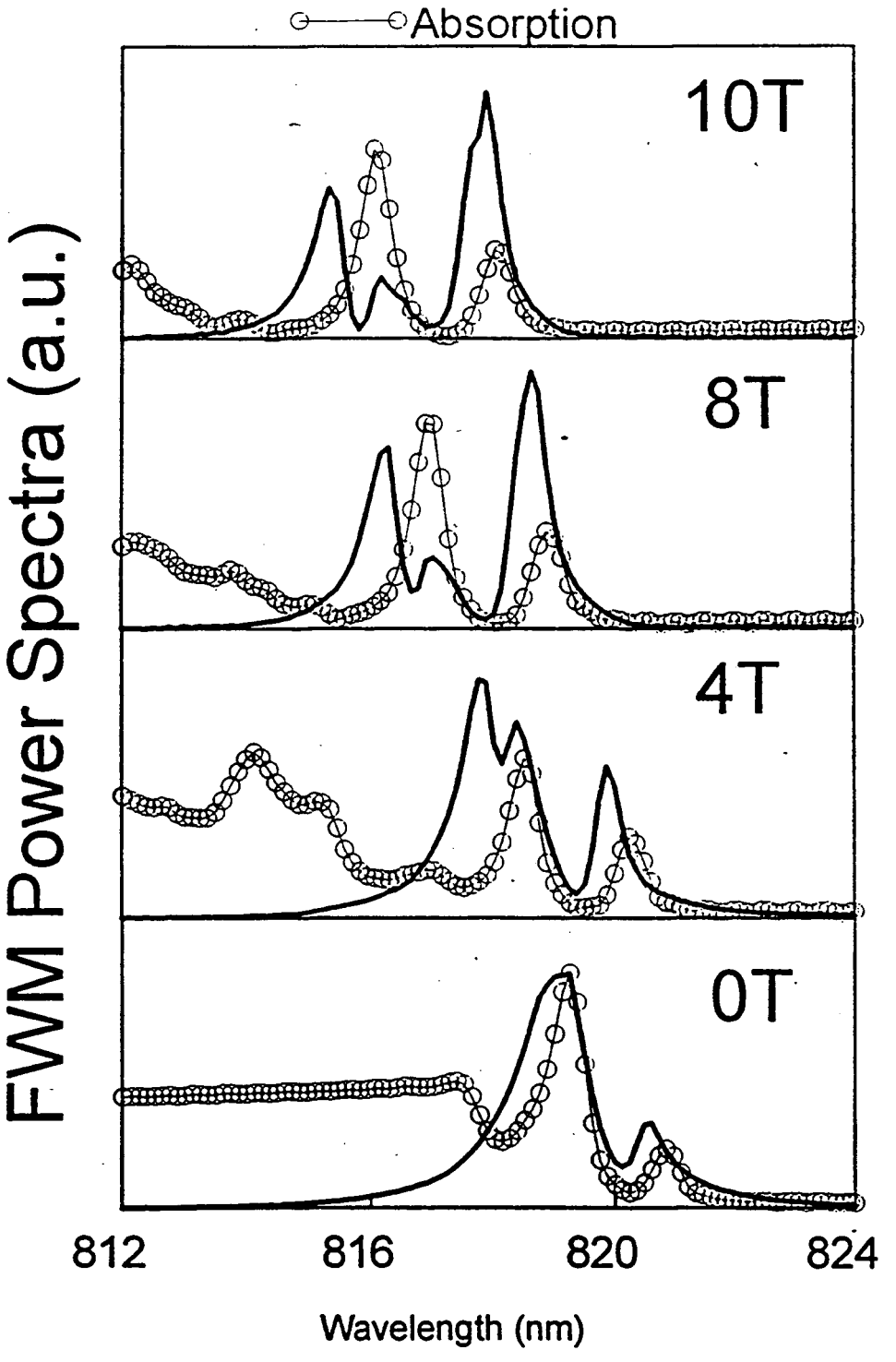
FIGURES

FIG.1: Top: linear absorption spectra of a high-quality GaAs sample at $T = 1.6\,\mathrm{K}$ and $B = 8\,\mathrm{T}$. Bottom: FWW spectra measured at the same field for excitation resonant with the lh and hh excitons at $N = 2 \times 10^{16}\,\mathrm{cm}^{-3}$ with σ^--polarized light. Inset: Two-pulse FWM configuration.

FIG.2: Comparison of the σ^--linear absorption and σ^-/σ^+-FWM spectra at $B = 0, 4, 8$, and $10\,\mathrm{T}$; $T = 1.6\,\mathrm{K}$, $\Delta t = 0$, and excitation density $N = 3 \times 10^{16}\,\mathrm{cm}^{-3}$.


Absorption
σ+
σ-
1.49
1.5
1.51
1.52
1.53
1.54
1.55
1.56
Energy (eV)


FWM Intensity
Δt= -50fs
Δt= -530fs
1.49
1.5
1.51
1.52
1.53
1.54
1.55
1.56
Energy (eV)


Absorption
10T
8T
4T
0T
FWM Power Spectra (a.u.)
812
816
820
824
Wavelength (nm)

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