Rabi Flopping in Semiconductors

S. T. Cundiff, A. Knorr, J. Feldmann, S. W. Koch, and E. O. Göbel
Fachbereich Physik und Wissenschaftliches Zentrum für Materialwissenschaften, Philipps Universität-Marburg, Renthof 5, D-35032 Marburg, Germany

H. Nickel
Deutsche Bundespost Telekom Fernmeldetechnisches Zentralamt, D-64295 Darmstadt, Germany
(Received 14 March 1994)

By examining the interaction of two copropagating ultrafast optical pulses in a semiconductor multiple quantum well, we experimentally determine the temporal dependence of the induced polarization. Based on this technique we observe that the optically induced density goes through a maximum at sufficiently high excitation intensity. Microscopic calculations show that the observed phenomena are a manifestation of Rabi flopping in semiconductors.

PACS numbers: 78.47.+p, 42.50.Md

Laser excited intrinsic excitations in a semiconductor provide a prototype for an interacting many-body system and, in particular, the coherent phenomena of the intrinsic excitations can yield extensive insight into the underlying many-body physics. The intrinsic excitations, however, display very short dephasing times, on the order of hundreds of femtoseconds or less, making the use of laser pulses with similar temporal durations necessary. Now that such pulses are routinely available, many of the optical measurements that were performed in atomic or molecular systems can be performed in semiconductors [1]. However, due to the many-body nature of a laser excited semiconductor, it is not *a priori* certain that coherent phenomena, such as free polarization decay, photon echo, quantum beats, self-induced transparency, etc., should even be observable, and if so whether they will be modified in a many-body system as compared to atoms or molecules. Consequently, if they are observed, any such modifications provide a powerful tool for studying the many-body interactions themselves. One coherent phenomenon that is theoretically predicted to occur in a semiconductor with many-body modifications [2], but that has not been directly experimentally observed, is the Rabi "flopping" of the optically induced excitation density, i.e., the induced excitation density is driven through a maximum by the incident field.

The oscillation of a two level system between the ground and excited states in the presence of a strong resonant driving field, often called transient nutation or Rabi flopping, is a basic quantum mechanical effect and a textbook topic today [3]. It was first treated by Rabi in the context of molecular beam magnetic resonance experiments [4] where it was also observed [5]. Later it was observed in magnetic resonance experiments in bulk material [6]. With the advent of the laser many of the resonance effects first studied in magnetic experiments were reproduced in optical experiments on atomic and molecular systems, including Rabi flopping [7]. The observation of density flops in semiconductors has been hindered by not only the rapid dephasing, but also by the fact the dephasing times are even shorter at elevated densities. And, to make matters worse in semiconductors, employing shorter pulses increases the hot carrier density, also increasing the dephasing. As a consequence, Rabi flopping in semiconductors has not been previously reported.

An important aspect of Rabi flopping is that it is a manifestation of the coupling between the optically induced coherence and the induced excitation density. As a consequence, it is sensitive to aspects of many-body phenomena that are not apparent in previously reported experiments in semiconductors, which have primarily focused only on the decay of the optically induced coherence (e.g., four wave mixing) [8]. For example, the theoretically expected enhancement of the Rabi flopping frequency by the internal field arising from many-body effects [2] is not observable in an experiment that is sensitive to the decay of the coherence alone.

The experimental technique presented here for observing density flops is based on measuring the modification of the shape of a femtosecond pulse that has propagated through a thin semiconductor film, similarly to the first studies in molecules [7]. However, because the effect is weaker and on a femtosecond time scale, more elaborate techniques are necessary. The pulse shape modification is observed via cross correlation with a reference pulse, as in experiments which have observed linear pulse propagation effects [9,10], however, this alone is not sufficient. The critical ingredient is the comparison of the pulse modification after propagation through an unexcited sample to that in a weakly excited sample. This is achieved by a weak copropagating prepulse, which is chopped, and performing differential detection via a lock-in amplifier [as depicted schematically in the inset in Fig. 1(a)].

To show that this technique is sensitive to the polarization (proportional to the temporal derivative of the excitation density), let us consider the cross-correlation signal detected by the lock-in, which is the difference between

0031-9007/94/73(8)/1178(4)$06.00

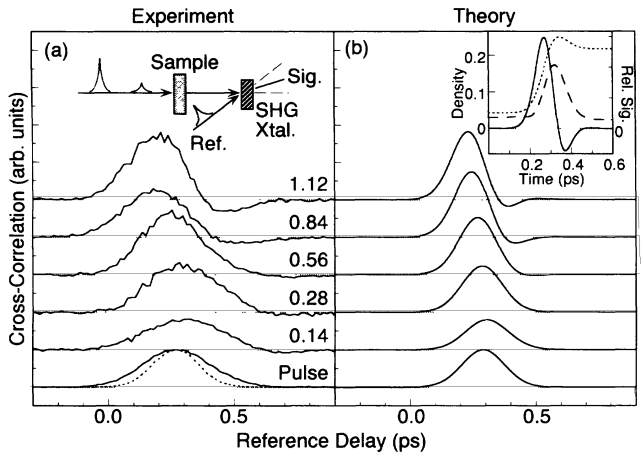

FIG. 1. Experimental (a) and theoretical (b) differential cross-correlation signal as a function of reference pulse delay and pulse intensity (traces offset vertically for clarity, zero is marked by dashed line and indicated intensities in GW/cm^2 apply to both experiment and theory). The bottom trace is the pulse cross correlation for reference [dotted line in (a) shows pulse without propagation through sample]. Inset in (a) shows experimental geometry. Inset in (b) shows the signal (solid line), calculated density with dephasing (dotted line) and without dephasing (dashed lined) as a function of time for a pulse intensity of 0.84 GW/cm^2.

pulse propagation with and without a prepulse, i.e.,

$$I_{\rm xc}(\tau) \propto \int I_d(t) I_r(t-\tau)\,dt - \int I_s(t) I_r(t-\tau)\,dt$$
$$= \int [I_d(t) - I_s(t)] I_r(t-\tau)\,dt\,,$$

where I_d (I_s) is the transmitted pulse intensity with (without) a prepulse and I_r is the intensity of the reference pulse. In turn the difference

$$I_{\rm sig} = I_d - I_s \propto |E + P_d/\varepsilon|^2 - |E + P_s/\varepsilon|^2$$
$$= (EP_d^* - EP_s^* + {\rm c.c.})/\varepsilon + (|P_d|^2 - |P_s|^2)/\varepsilon^2,$$

where E is the electric field of the incident pulse, P_s (P_d) the polarization induced by the pulse without (with) a prepulse, and ε the dielectric constant. For $E \gg P$ the last two terms can be ignored and only the absorptive polarization component need be considered.

To proceed further, we discuss the basic idea using the example of a two level system before proceeding to a more realistic analysis for semiconductors, where we must resort to numerical techniques. The Rabi solution to the optical Bloch equations (OBE) for a strongly driven two level system, where the driving field is constant and switched on at $t = 0$ (i.e., a square pulse), is [11]

$$P \propto n_0 \frac{\omega_R}{\Omega_R} \sin(\Omega_R t), \quad \Omega_R = \sqrt{\Delta^2 + \omega_R^2}, \quad \omega_R = \frac{\mu E}{\hbar},$$

if there is no coherence present in the sample at $t = 0$, and where Δ is the frequency detuning between the field and resonance, μ the dipole moment, E the electric field, and n_0 the inversion at $t = 0$. The inversion is $n_0 = -1$ for a system in the ground state and increases to $+1$ for a completely inverted system. If the effect of the prepulse is to induce a small excitation which increases n_0 by δ, then

$$I_{\rm sig} \propto (\delta E \omega_R/\Omega_R)\sin(\Omega_R t) + {\rm c.c.}$$

The corresponding temporal behavior of the inversion is given by [11]

$$n = n_0[\Delta^2 + \omega_R^2 \cos(\Omega_R t)]/\Omega_R^2$$

from which we see that a zero crossing in the signal occurs at the point at which the inversion reaches a maximum (for an unexcited or weakly excited system $n_0 < 0$). Therefore we conclude that (1) a zero crossing that appears for sufficiently high intensity is a signature of Rabi flopping, (2) the zero crossing shifts toward decreasing time with increasing pulse area, and (3) it is independent of prepulse intensity.

This result can be understood intuitively by considering the limiting case where the prepulse drives the system to transparency ($n = 0$), and the square primary pulse causes exactly one Rabi flop (a 2π pulse). In the absence of the prepulse the system undergoes a Rabi flop, and during the first half of the pulse it absorbs energy from the pulse as it goes from the ground state to completely inverted, after which it returns energy to the pulse as it is driven back to the ground state, i.e., the second half of the pulse sees gain, analogously to self-induced transparency [11]. If the prepulse incoherently drives the system to transparency, neither of these occur; the pulse simply propagates through the sample unimpeded. Hence, in the presence of the prepulse, the first half of the pulse is stronger, because the absorption is missing, and the second half weaker, because the gain is missing. Consequently, when the difference between the transmitted pulses with and without a prepulse is taken, a positive difference occurs for the first half of the pulse and a negative one for the second half. This picture is a gross simplification, the first pulse does not drive the system to transparency and the pulses are not square, but it does provide insight into the technique.

To observe Rabi flopping in semiconductors we employ this technique on an $In_{0.08}Ga_{0.92}As/GaAs$ 20 period multiple quantum well, with 10 nm wells and 40 nm barriers grown on a GaAs substrate. It is held at 5.5 K in helium vapor and displays a strong 1s exciton resonance at 1.454 eV. The GaAs substrate is not removed as it is transparent at this energy. The sample is described in more detail elsewhere [12]. A self-mode-locked Ti:sapphire laser produces the incident pulses, which are initially 100–110 fs duration and not completely bandwidth limited. Because of glass in the optical path the pulses incident on the sample are chirped and have a duration of 140–150 fs and an 18–20 meV full width at half maximum bandwidth. After passage through the sample the pulses are further stretched by the strongly dispersive GaAs substrate.

The experimental excitation conditions are chosen so as to emphasize the desired phenomena. For a pulse resonant with a strongly absorbing transition, linear effects, which are sensitive to the dephasing rate, alone can result in strong distortion of the transmitted pulse [10]. Additionally nonlinear propagation effects can complicate interpretation. To minimize these effects the peak of the laser spectrum is tuned 9 meV below the $1s$ exciton resonance for all results presented below. The low intensity absorption of the pulse is $\alpha L = 0.15$. The free polarization decay due to the prepulse is observable and yields an estimated dephasing time of 300 fs (this is already substantially shorter than the low density value [12]). Because the pulse separation is 1.5 psec in the experiment, coherent interaction between the pulses is not important. A comparison between single and double pulse propagation indicates that nonlinear propagation effects are not experimentally significant.

In Fig. 1(a) the measured differential cross-correlation signal as a function of reference delay is shown. The figure shows the cross-correlation signal which corresponds to the modification of the second pulse due to the prepulse and positive signal indicates an increase in the detected intensity when the prepulse is present. Zero reference delay corresponds to the arrival of a pulse tuned well below the resonance. For reference the bottom trace is the cross correlation of the pulse propagating through the sample alone (the dotted line is the cross correlation for no sample, demonstrating the broadening due to the substrate). Successive traces from bottom to top correspond to increasing pulse intensity, varying from 0.14 to 1.12 GW/cm^2 to within 10% including all reflection losses. At 0.14 GW/cm^2 the signal is essentially the same as the pulse, although the peak is slightly delayed. There is a small negative signal after the pulse that is due to the suppression of the free decay due to the enhancement of the dephasing rate caused by the prepulse excitation. Once the intensity is increased to 0.56 GW/cm^2 the signal becomes asymmetric, at 0.84 GW/cm^2 a zero crossing during the pulse appears, which shifts toward the center of the pulse at 1.12 GW/cm^2. *This appearance of a zero crossing above a critical intensity that shifts with a further increase in intensity is exactly the behavior expected for Rabi flopping.*

For a two level system the correspondence between a zero crossing in the signal and a density flop is strict. However, in order to verify that the zero crossings we observe in semiconductors are indeed a signature of Rabi flopping, we must perform microscopic calculations. In a microscopic semiconductor theory the OBE must be replaced with semiconductor Bloch equations (SBE), which for a two-band semiconductor dipole coupled to an external field $\hat{E}(z,t)$, can be written [13]

$$\frac{\partial P_{\mathbf{k}}}{\partial t} = -i(e_{e,R} + e_{h,R})P_{\mathbf{k}} - i(f_{e,\mathbf{k}} + f_{h,\mathbf{k}} - 1)\omega_{R,\mathbf{k}} + \left.\frac{\partial P_{\mathbf{k}}}{\partial t}\right|_{\text{col}},$$

$$\frac{\partial f_{l,\mathbf{k}}}{\partial t} = (iP_{\mathbf{k}}^{*}\omega_{R,\mathbf{k}} + \text{c.c.}) + \left.\frac{\partial f_{l,\mathbf{k}}}{\partial t}\right|_{\text{col}}, \quad l = e,h,$$

where

$$e_{l,k} = \varepsilon_{l,k} + \frac{1}{\hbar}\sum_{\mathbf{q}} V_{|\mathbf{k}-\mathbf{q}|} f_{l,\mathbf{q}} + \Sigma_{\text{CH}}, \quad l = e,h,$$

$$\omega_{R,\mathbf{k}} = \frac{1}{\hbar}\left[d_{cv}E + \sum_{\mathbf{q}\neq\mathbf{k}} V_{|\mathbf{k}-\mathbf{q}|}P_{\mathbf{q}}\right],$$

$\varepsilon_{l,k}$ is the zero density energy dispersion, d_{cv} the dipole matrix element, E the envelope of the applied electric field, $\hat{E}(z,t) = E(z,t)\exp[-i\omega_0 t + ik_0 z]$, $V_{\mathbf{k}}$ the **k**th Fourier component of the screened Coulomb potential, and Σ_{CH} the Coulomb hole. Relaxation is treated by the phenomenological collision terms $\partial P_{\mathbf{k}}/\partial t|_{\text{col}}$ and $\partial f_{l,\mathbf{k}}/\partial t|_{\text{col}}$. We assume a constant polarization dephasing time and treat the carrier intraband collisions in the relaxation rate approximation [13]. The propagation of the pulse through the sample is described by the reduced wave equation

$$\frac{\partial E(\eta,\xi)}{\partial \xi} = \frac{i\mu_0\omega_0^2}{k_0 V}\sum_{\mathbf{k}} P_{\mathbf{k}}(\eta,\xi)$$

$$(\eta = z - t/v_g,\ \xi = z),$$

where v_g is the group velocity. To include possible propagation effects the coupled Maxwell-SBE are solved numerically for two copropagating pulses.

In Fig. 1(b) the numerical results are shown for conditions corresponding to the experiment. The numerical results show behavior identical to the experimental results. As a function of intensity, they change from being symmetric and slightly delayed to showing a zero crossing, which shifts with increasing intensity. For these calculations a dephasing time of 200 fsec was used to include further enhancement of the dephasing due to the stronger second pulse, consistent with the upper limit obtained from the observed free decay following the first pulse. Note that the second zero crossing at the highest intensity and following weak positive signal in the numerical results are a manifestation of the nonlinearity of the SBE. Because the frequency of oscillation between ground and excited states depends on the detuning, and hence so does the position of the zero crossing in the experiment, not only the pulse width, but also the spectral position and content of the incident fields employed in the experiment must be closely reproduced in the calculation. Therefore the calculations include the chirp on the incident pulses. The additional chirp due to the GaAs substrate after the sample is not included as it does not influence the physics, however, it does result in additional stretching in the experimental results compared to the theoretical ones.

To show that the density does go through a maximum, we show the calculated density and signal as a function of time in the inset to Fig. 1(b). A density maximum corresponding to the zero crossing is present. The fact that the density decreases only ~15% after the maximum is due to the high dephasing rate, and to show this we also plot the evolution without dephasing for comparison. In this case the residual density is primarily due to many-body effects [2]. 0.84 GW/cm^2 corresponds to a pulse area of $\sim 1.3\pi$ for a two level system, which in the presence of the dephasing results in such a weak density maximum that no zero crossing is observable for a two level system. We therefore conclude that the oscillation frequency enhancement due to the internal field arising from many-body effects is necessary for the zero crossing to be present at these intensities [14].

For these theoretical results, which correspond to our best estimate of the experimental conditions, the density flop is clearly identified as Rabi flopping. Generally, for off-resonant excitation, nonmonotonic density dynamics can be observed [15] due to adiabatic following. The transition between Rabi flopping and adiabatic following is sensitive to both temporal and spectral pulse widths. Under our experimental conditions the pulse spectrum shows strong overlap with the exciton resonance, despite the detuning, as a consequence of the chirp. Therefore both adiabatic following and Rabi flopping contribute to the experimental results. For all parametric excursions in the calculations that are still consistent with the experimental conditions, the primary contribution is clearly due to Rabi flopping.

To verify that the zero crossing also fulfills the third requirement for Rabi flopping, namely that it is independent of the prepulse intensity, we present the experimental result across an order of magnitude variation in prepulse intensity in Fig. 2(a) and in Fig. 2(b) the corresponding calculations. At low intensity the zero crossing is essentially independent of prepulse intensity, at the highest intensity it shifts to later times. This shifting to later times can be explained by the increased dephasing rate due to the higher prepulse intensity, and an increase appears in the calculation if the dephasing rate is increased [dashed line in (b), note that the prepulse always increases the dephasing rate compared to an unexcited crystal, however, it does not affect the signal until it becomes significant in comparison to the dephasing due to the density excited by the primary pulse]. These results are not only consistent with the aforementioned expectation, but they also eliminate the possibility that nonlinear propagation effects are important. In particular, a bleaching of the exciton resonance due to the prepulse resulting in increased transmission and modification of the group velocity, could produce a zero crossing, however, the group velocity then depends on the first pulse intensity and hence so would any zero crossing.

Finally, we note that by minimizing the chirp on the incident pulse we observe indications of a (possibly partial) second flop. In this case the higher flopping frequency makes the time resolution of the cross correlation the limiting factor and more detailed analysis necessary.

In conclusion, we have presented the results of an experimental technique that allows us to observe density flops in semiconductors. Theoretical calculations identify the density variations as Rabi flops. The results clarify the conditions in a semiconductor under which this class of coherent experiment is possible and provide insight

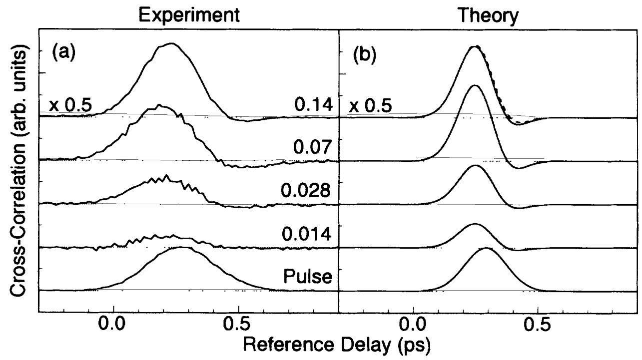

FIG. 2. As Fig. 1, except as a function of prepulse intensity. Dashed line in (b) shows effect of higher dephasing rate.

into the feasibility of other related phenomena (e.g., self-induced transparency), which in turn will provide further, otherwise inaccessible, insight into the influence of many-body interactions on the coupling between the induced coherence and excitation.

The work was supported through the Leibniz- and Gerhard Hess-Förderpreise. CPU time from HRLZ Jülich is also acknowledged. One of the authors (S. T. C.) acknowledges support from the von Humboldt Foundation. The authors thank M. Preis for his technical assistance.

[1] N. Peyghambarian, S. W. Koch, and A. Mysyrowicz, *Introduction to Semiconductor Optics* (Prentice Hall, Englewood Cliffs, 1993).
[2] R. Binder *et al.*, Phys. Rev. Lett. **65**, 899 (1990).
[3] C. Cohen-Tannoudji, B. Diu, and F. Laloë, *Quantum Mechanics* (Wiley, New York, 1977).
[4] I. I. Rabi, Phys. Rev. **51**, 652 (1937).
[5] I. I. Rabi, S. Milliman, P. Kusch, and J. R. Zacharias, Phys. Rev. **55**, 526 (1939).
[6] H. C. Torrey, Phys. Rev. **76**, 1059 (1949).
[7] G. B. Hocker and C. L. Tang, Phys. Rev. Lett. **21**, 591 (1968).
[8] S. Weiss *et al.*, Phys. Rev. Lett. **69**, 2685 (1992); D.-S. Kim *et al.*, Phys. Rev. Lett. **69**, 2725 (1992).
[9] R. G. Ulbrich and G. W. Fehrenbach, Phys. Rev. Lett. **43**, 963 (1979); S. Chu and S. Wong, Phys. Rev. Lett. **48**, 738 (1982); J. Hegarty, Phys. Rev. B **25**, 4324 (1982); K. Ogawa, T. Katsuyama, and H. Nakamura, Phys. Rev. Lett. **64**, 798 (1990).
[10] D.-S. Kim *et. al.*, Phys. Rev. B **48**, 17 902 (1993).
[11] L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975).
[12] J. Feldmann *et al.*, Phys. Rev. Lett. **70**, 3027 (1993).
[13] H. Haug and S. W. Koch, *Quantum Theory of the Optical and Electronic Properties of Semiconductors* (World Scientific, Singapore, 1993), 2nd ed.
[14] T. Östreich and A. Knorr, Phys. Rev. B **48**, 17 811 (1993); A. Knorr *et al.*, Phys. Rev. B **49**, 14 024 (1994).
[15] P. A. Harten *et. al.*, Phys. Rev. Lett. **69**, 852 (1992).

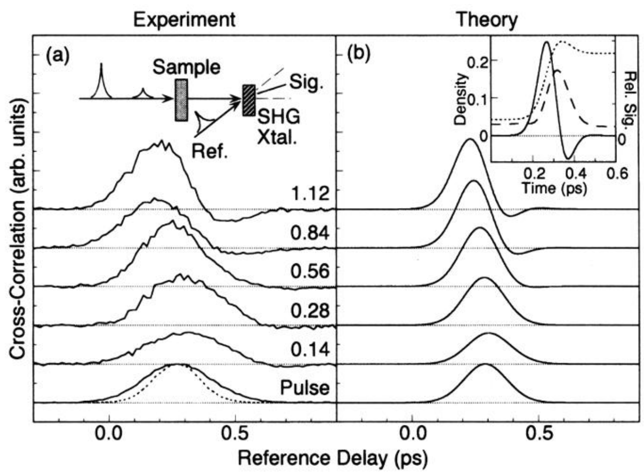

FIG. 1. Experimental (a) and theoretical (b) differential cross-correlation signal as a function of reference pulse delay and pulse intensity (traces offset vertically for clarity, zero is marked by dashed line and indicated intensities in GW/cm^2 apply to both experiment and theory). The bottom trace is the pulse cross correlation for reference [dotted line in (a) shows pulse without propagation through sample]. Inset in (a) shows experimental geometry. Inset in (b) shows the signal (solid line), calculated density with dephasing (dotted line) and without dephasing (dashed lined) as a function of time for a pulse intensity of 0.84 GW/cm^2.