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13. ABSTRACT (Maximum 200 words) This report focuses on the generation of coherent phonon fields using ultrafast optical pulses and theoretical work on coherent control and Bose-Einstein condensation of excitons. Our results illustrate the concept of phonon field entanglement and establish the relationship between phonon generation in opaque substances and resonant Raman scattering. We consider specifically coherent modes in LaAlO ₃ , squeezed quantum-entangled phonons in KTaO ₃ , and thermally-entangled transverse optical-acoustical modes in SrTiO ₃ . The resonant work covers the E ₂ ' - resonance of antimony and the band gap of GaSe. Our work on condensates centers on light emission from optically-active excitons in quantum-well structures. We show that a coherent electromagnetic field is emitted and that condensation in a two-dimensional system can take place at finite temperatures. The bosonic description is extended to consider a weak disorder potential. We find that the collective state is a Glauber coherent state, substantially different from that of few-level models. The theory is used to explain transient linear optical experiments involving resonantly excited excitons in GaAs structures. We consider particularly coherent control of the exciton density and spin and resonant Rayleigh scattering. Our quantum mechanical approach for light emission shows that excitonic secondary emission has a coherent component, in agreement with experiments.				
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CONTROL OF ELECTRON COHERENCES WITH COHERENT PHONONS

FINAL REPORT

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I. FOREWORD

In this report, we present a succinct account of research findings for projects supported by ARO Contract No. DAAH04-96-1-0183 [Amount: \$260,000 (includes \$15,000 university cost sharing contribution) for the period July 1, 1996 through December 31, 1999]. The research divides into two components dealing with the generation of coherent phonon fields using ultrafast optical pulses and theoretical work on optical coherent control and Bose-Einstein condensation of excitons. The latter work was performed in collaboration with colleagues from the Universidad Autónoma de Madrid, Spain. Other than the PI, the phonon work involved two graduate students. Some of the ultrafast science projects benefited from the participation of Dr. Thomas Albrecht, a Feodor-Lynen Fellow from the University of Marburg, Germany and Dr. Alka Ingale, a Boyscast Fellow from the Centre for Advanced Technology, Indore, India.

II. TIME-DOMAIN COHERENT RAMAN SCATTERING

In this section, we present a brief introduction to phonon generation mechanisms based on impulsive stimulated Raman scattering (ISRS) [1-24]. For phonons, light pulses generate both coherent and squeezed states. An in-depth treatment of coherent phonons can be found in the reviews available in the literature [25-29] including a recent one by the PI where opaque systems are thoroughly discussed [29].

Ultrafast measurements are performed with a standard pump-probe setup. The signal of interest is the transmitted (or reflected) intensity of the probe beam as a function of the time delay, as measured by the relative distance between the two pulses. In transparent materials, the generation of phonon coherences originates in the modulation of the refractive index n or, alternatively, the electronic susceptibility $\chi = (n^2-1)/4\pi$, by the ion motion. To first order in the phonon amplitude Q_q , we have that $\delta n \approx 2\pi[\partial\chi/\partial Q_q]Q_q/n$; q is the phonon wavevector. Because the wavelength of the light is much larger than the lattice parameter, only modes with $q \approx 0$ can couple to light. The resulting change in electromagnetic energy density is

$$\delta U^{(1)} = \frac{1}{2} \delta\chi^{(1)} |E_0|^2 = \frac{|E_0(t)|^2}{2} \left(\frac{\partial\chi}{\partial Q} \right)_{Q_{q=0}} \quad (1)$$

where $E_0(t)$ is the magnitude of the pump electric field. Since $\delta U^{(1)} \propto Q_{q=0}$, Eq. (1) gives a *force density* $F^{(1)}$ acting on $Q_{q=0}$ which is proportional to the electric field intensity. This interaction is associated with coherent states. If we ignore phonon dispersion and dissipation, the classical equation of motion for the amplitude (as well as the quantum-mechanical expression for the expectation value) is

$$d^2 Q_{q=0} / dt^2 + \Omega^2 Q_{q=0} = |E_0(t)|^2 (\partial\chi / \partial Q) / 2 = F^{(1)} \quad (2)$$

which describes a driven harmonic oscillator of frequency Ω . Since the lattice motion is controlled by Fourier components of $F^{(1)}$ at $\approx \Omega$, the coupling is large only if the duration of the pulse τ_0 is small compared with Ω^{-1} . Note that, for $\tau_0 \ll \Omega^{-1}$, $Q_{q=0} \propto \sin(\Omega t)$. The properties of the coherent field are usually studied by monitoring the

scattering of the probe pulse by the time- and space-varying refractive index associated with $Q_{q=0}$ ($\delta n \propto Q_{q=0}$). If we consider the fact that the susceptibility is not a scalar but a tensor, the expression for the force is $F^{(1)} = \sum_{\mu\nu} (R_{\mu\nu} E_\mu E_\nu) / 2$. Here, E_μ denotes a component of the pump field, $R_{\mu\nu} \approx \partial \chi_{\mu\nu} / \partial Q$ is the first-order Raman scattering tensor [29] and $\chi_{\mu\nu}$ is the linear susceptibility. This relationship between the driving force and $R_{\mu\nu}$ is the reason why the pulsed generation of coherent modes is commonly referred to as ISRS. In this context, we recall that the spontaneous cross-section is proportional to $|R_{\mu\nu}|^2$; hence, ISRS and conventional Raman spectra are closely related. These considerations apply strictly to transparent substances but, as discussed in [29] and supported by our resonant impulsive results [23], they can be extended to cover opaque materials as well.

The generation of squeezed states relies on the second-order term [30]

$$\delta U^{(2)} = |E_0(t)|^2 \sum_k (\partial^2 \chi / \partial Q_k^2) Q_k^2 / 2. \quad (3)$$

giving $\delta n \propto \sum (Q_k)^2$ and a force $\propto Q_k$. The spontaneous process associated with Eq. (3) is two-phonon Raman scattering involving pairs of modes at $\pm \mathbf{k}$. Depending on whether we can consider standing or running waves, this coupling describes, respectively, a time-varying change in the frequency of a single mode or $\pm \mathbf{k}$ mixing.

For a single oscillator, the time-dependent Schrödinger equation can be solved exactly in the limit where the width of the laser pulse is very small compared with the oscillator period. Then, we can approximate $|E_0(t)|^2 \propto \delta(t)$ and we have that

$$\Psi_1(0^+) = \exp(i\alpha Q) \Psi(0^-) \quad \text{and} \quad \Psi_2(0^+) = \exp(i\beta Q^2) \Psi(0^-) \quad (4)$$

for $\delta U^{(1)}$ and $\delta U^{(2)}$; α and β are constants. In the case where $\Psi(0^-)$ is the ground state of the oscillator, Ψ_1 and Ψ_2 describe, respectively, states closely related to the well-known Glauber's coherent state and the so-called two-photon coherent state of quantum optics [31-32]. As in the classical case, the coherent state Ψ_1 exhibits an instantaneous change in the expectation value of the momentum $\langle p \rangle$ while $\langle p^2 \rangle$ changes abruptly for the squeezed state Ψ_2 . It should be emphasized that neither state is stationary. In the coherent case, $\langle p \rangle$ oscillates back and forth following the classical equation of motion but $\langle p^2 \rangle$ does not change. The opposite applies to the Ψ_2 for which $\langle p \rangle = 0$ while $\langle p^2 \rangle$ oscillates at twice the phonon frequency remaining below the value for the ground state for half a cycle.

II.1 Survey of Results

In this section, we review our ISRS experiments. Our results illustrate the concept of (quantum and thermal) field entanglement for phonons and establish the relationship between resonant and non-resonant-ISRS. These studies were supported in part by U. S. Army Research Office, Physics Division. Additional funds were provided by the NSF through the Center for Ultrafast Optical Science and by the von Humboldt Foundation of Germany through the Feodor-Lynen program.

The resonant work covers the E'_2 - resonance of antimony [23,33] and an investigation of GaSe using impulsive excitation resonant with the band gap [33]. We also discuss squeezed phonons in KTaO_3 , representing quantum entanglement between states at $\pm \mathbf{k}$ of the same branch [30], and (thermally-) entangled transverse optical-acoustical modes in SrTiO_3 [34]. Finally, we review our work on coherent modes in LaAlO_3 [12]. The latter three compounds belong to a large family of wide-gap perovskites which commonly undergo phase transformations associated with the softening of a phonon branch in the neighborhood of a certain point of the Brillouin zone [35]. While these compounds share in common the high-temperature cubic structure, their low-temperature form varies widely [35]. Extensive soft phonon investigations of perovskite transitions have been performed in the past twenty years or so using spontaneous RS [35] and, more recently, using ISRS [17-21]. With few exceptions, the latter studies center on ferroelectric compounds, such as PbTiO_3 [17] and LiTaO_3 [21], for which the relevant mode, associated with the Γ -point of the cubic structure, is both Raman and infrared active at low temperatures. Since ISRS probes wavevectors in the range where infrared modes couple strongly to light (as in forward RS) [29], the data are dominated by polariton effects [29].

Our work on LaAlO_3 was motivated primarily by the fact that its low-temperature form retains the inversion symmetry of the cubic phase. As a result, the soft mode (originally at the R -point) splits below the transformation into an A_{1g} singlet and an E_g doublet which are active in Raman but not in the infrared. Thus, there are no polariton effects for ISRS in LaAlO_3 . Results for the E_g -mode are reproduced in Fig. 1. The phase transition occurs at $T_C \approx 750$ K. With increasing temperature, the ISRS data show a decrease in frequency and an increase in the decay rate, in excellent agreement with spontaneous RS spectra. This, as well as a comparison between values from the two sets of data for the relative ratio between the A_{1g} (not shown) and E_g intensities confirm the Raman origin of the phonon-generation process. The absolute value of the ratio between the coherent vibration amplitude and the pulse areal density can be gained from measurements of the relative transmission, $\Delta T/T$ [12]. In LaAlO_3 , we find $0.6\text{-}6 \text{ m}\mathring{\text{A}}/(\text{Jm}^{-2})$ which, for a

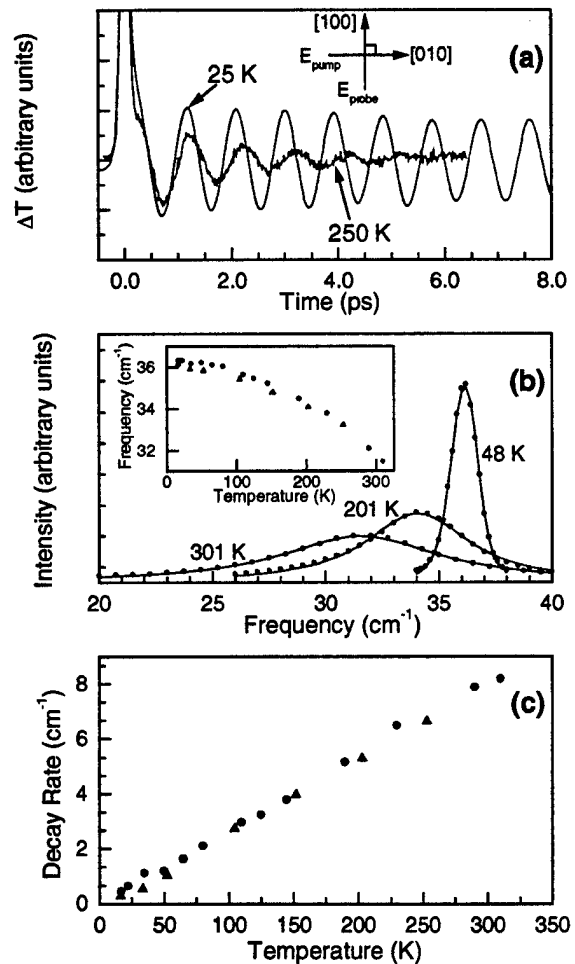


FIGURE 1 - Data from LaAlO_3 . (a) Coherent E_g oscillations obtained in transmission with pulses at 815 nm. (b) Spontaneous Raman spectra showing E_g scattering at 647.1 nm (laser wavelength); solid lines are Lorentzian fits. The inset compares time-domain (red triangles) and spontaneous RS (green circles) results on the T -dependence of $\Omega(E_g)$. (c) $\Gamma(E_g)$ vs. T .

beam focused to a 50 μm -diameter spot, a repetition rate of 80 MHz and an average power of 100 mW, translates into average displacements in the range 50-500 $\mu\text{\AA}$ [12].

Unlike LaAlO_3 , KTaO_3 does not show any phase transition and remains cubic down to, essentially, $T = 0$. Similar to SrTiO_3 , however, the low-frequency dielectric constant exhibits a plateau below ≈ 20 K at a value exceeding 10^4 and a concomitant pronounced softening of an infrared-active mode at the center of the Brillouin zone [36]. Both KTaO_3 and SrTiO_3 are examples of quantum paraelectricity [36-37]. The large value of the response reflects an incipient transformation into a ferroelectric phase, which does not take place due to quantum-fluctuations [37]. KTaO_3 is nearly the ideal choice for studies of squeezing because it combines low-frequency modes (which are advantageous in that they can be probed using pulses that are not too narrow) with the cubic perovskite structure which does not allow for first-order scattering. Hence, $\delta U^{(1)} \equiv 0$ [Eq. (1)] exactly and, therefore, the leading term describing the coupling of light to vibrations is the squeezing potential $\delta U^{(2)}$ [Eq. (3)]. Time-domain results for KTaO_3 at $T = 10$ K are shown in Fig. 2. The Fourier transform $F_{\text{FT}}(\Omega) = \int (\Delta T/T) \cos(\Omega\tau) d\tau$ (Fig. 2b) is dominated by

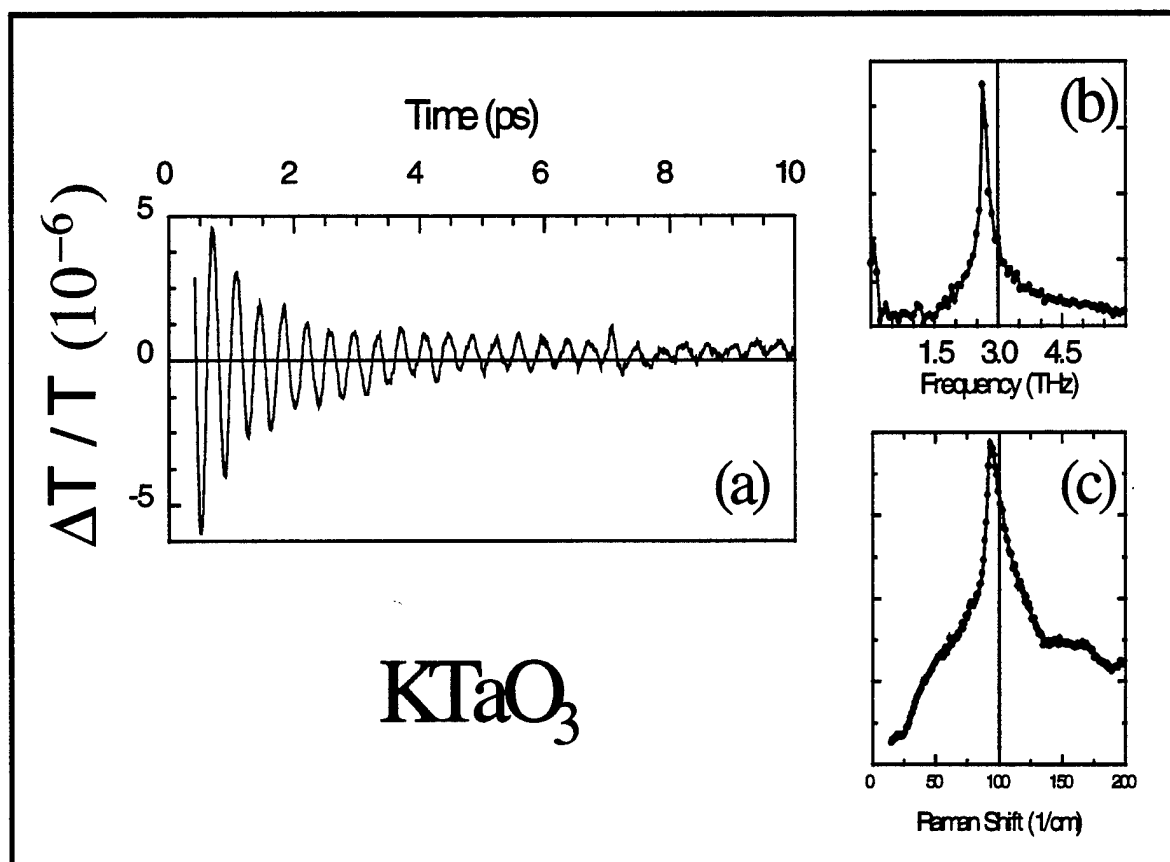


FIGURE 2. (a) Normalized transmitted intensity of the probe pulse as a function of the delay for KTaO_3 at $T = 10$ K. The symmetry configuration is A_{1g} . (b) Fourier transform of the time-domain data. (c) Weighted second-order RS cross section (see text) obtained at 810.0 nm.

a narrow peak, strongly dependent on temperature, that appears very close to twice the frequency of the transverse acoustic (TA) mode at the X-point of the Brillouin zone, as measured by neutron scattering. This, as well as the comparison between the time-domain and the spontaneous RS measurements (Fig. 2c) indicate that the oscillations in Fig. 2a are due to the 2TA overtone, confirming the $\delta U^{(2)}$ nature of the light-phonon interaction. The sharpness and strength of the 2TA peak reflects to some extent the flatness of the phonon dispersion near the zone boundary. For a given irreducible component, it can be shown from previously derived expressions [30] that $F_{FT}(\Omega)$ is proportional to the second-order RS intensity divided by the weighting factor $C(\Omega) = (\hbar/2\Omega)[1 - \exp(-\hbar\Omega/k_B T)]^{-1}$. The comparison between Fig. 2b and 2c indicates that the theoretical prediction is in reasonable agreement with the experimental data. However, there are significant differences concerning the line-shape, particularly the width of the main feature, that are not well understood [30].

From the data in Fig. 2a and measurements of the absolute RS cross section, it is possible to obtain a quantitative estimate of the variance

$$\langle U^2(t) \rangle = \sum_l \frac{M(l)}{M_T} \langle u^2(l) \rangle = \sum_q \frac{\langle Q_q^2(t) \rangle}{NM_T} \quad (5)$$

which provides a measure of the squeezing in real space [30]. Here, $u(l)$ and $M(l)$ are, respectively, the deviation of equilibrium and the mass of the l th-atom in the unit cell, M_T is the mass of a single cell and N is the total number of

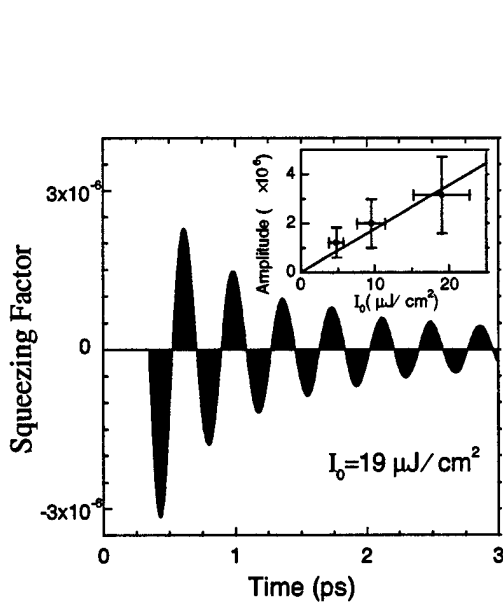


FIGURE 3. Experimental time dependence of the squeezing factor S for KTaO_3 . I_0 is the integrated pulse intensity. Inset: Amplitude of S vs. I_0 .

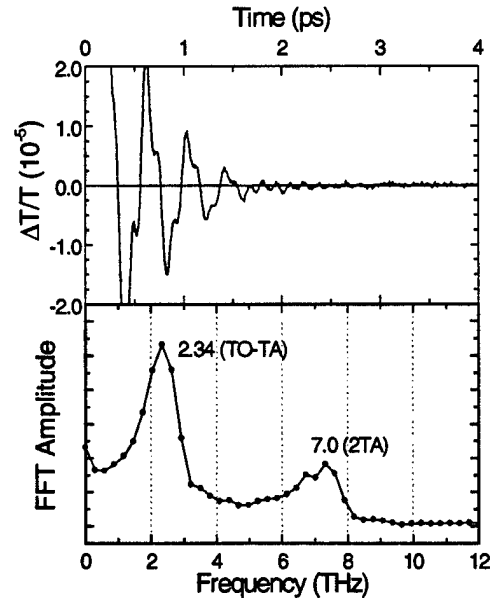


FIGURE 4. Time-resolved differential transmission in SrTiO_3 at 300K. The FFT amplitude shows the presence of two second-order phonon fields, one at 2.3 THz due to difference scattering and an oscillation at 7.3 THz due to sum scattering.

cells. In Fig. 3, we plot the squeezing factor $S = 1 - [\langle U^2(t) \rangle / \langle U^2(0) \rangle]^{1/2}$; $S > 0$ corresponds to values below the standard quantum limit [30].

ISRS data for SrTiO₃ at room temperature is shown in Fig. 4 [34]. Similar to KTaO₃, the main features are due to second-order scattering. The peak at lower frequencies is due to difference scattering and it is associated with the correlated motion of transverse optical (TO) and acoustic phonons. The high-frequency oscillation, labeled 2TA, reflects thermal squeezing involving $\pm k$ transverse acoustic modes. As for spontaneous scattering, the ISRS TO-TA can only occur in the presence of thermal phonons [34].

The relationship between vibrational coherences and spontaneous Raman scattering is well established only for transparent substances. Here, the pulses translate into an impulsive force giving $Q \propto \sin(\Omega t)$ when the pulse width is small compared with Ω^{-1} . For opaque materials, however, a unique process has so far not been identified [5,29]. One of the leading proposals is the mechanism known as *displacive excitation* of coherent phonons (DECP). The DECP model [5] provides a simple explanation for driving fully symmetric (*A*-type) modes, seemingly unrelated to Raman scattering. In the DECP picture, the equilibrium positions of the ions experience a sudden shift due to coupling with photoexcited carriers created by the optical pulse. Hence the force is step-like and, thus, $Q \propto \cos(\Omega t)$. Our studies on antimony [23] contain theoretical and experimental results which question the validity of the DECP interpretation.

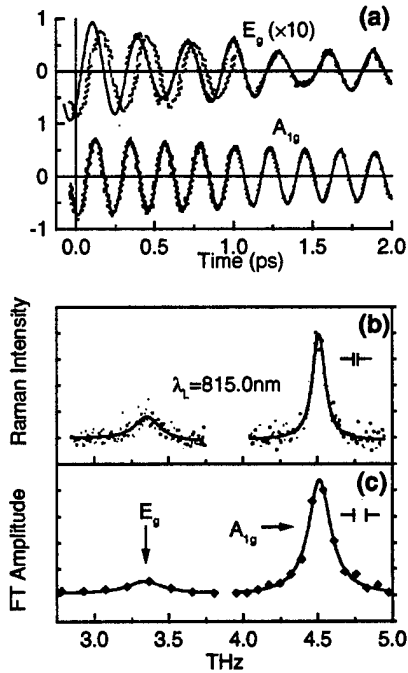


FIGURE 5. (a) Reflection geometry data in Sb. Fits to $\exp(-\Gamma t)\sin(\Omega t + \phi)$ are shown after removal of the background. Dashed curves are $\propto \cos(\Omega t)$ corresponding to the displacive limit. The bottom panel compares the RS intensity (b) with the FFT amplitude (c) gained from the traces in (a). Solid lines are Lorentzian fits.

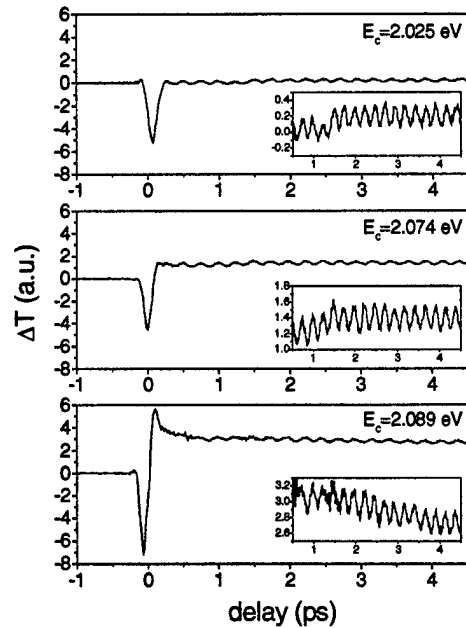


FIGURE 6. Time-domain traces for ϵ -GaSe at 77 K. Data show the dependence of the coherent phonon oscillations on the central energy of the laser pulse in the vicinity of the gap (polarization perpendicular to the *c*-axis).

The theoretical work shows that the DECP model can be obtained as a particular case of *resonant* coherent RS [23] and, therefore, that Raman scattering provides a unifying approach for describing light-induced motion of atoms of both impulsive and displacive character. The time-domain data, shown in Fig. 5, confirm these findings. Unlike previous studies [5], we find (i) that pulses drive not only the fully-symmetric A_{1g} but also the E_g Raman allowed mode (Fig.5 a), (ii) that there is a strong correlation between time-domain and RS intensities (contrast Figs. 5b and 5c), and (iii) that the phase of the oscillation is intermediate between that of purely displacive and impulsive dynamics (see fits in Fig. a). The comparison between results from [2], obtained at ≈ 2 eV, and our measurements at ≈ 1.5 eV reveal a crossover for the A_{1g} mode from purely displacive, at 2 eV, to mixed-behavior. This, and the fact that the E'_2 -resonance is centered at ≈ 2.15 eV, indicates that the displacive behavior relies on *two-band* contributions to the Raman process, in agreement with theoretical predictions [29].

To further test the relationship between spontaneous Raman and ISRS, we performed resonant impulsive measurements on ϵ -GaSe using an optical parametric amplifier [33]. In Fig. 6, we show data using pulses of central energy E_C in the vicinity of the direct exciton (at $E_X \approx 2.1$ eV). The oscillations are associated with the A_1 -symmetry mode at $\Omega \approx 4$ THz. The phonon contribution could be clearly distinguished from the electronic background for energies below $\sim E_X + \hbar\Omega$, but it becomes harder to separate above that value. The dependence of the phonon *amplitude* as a function of E_C (not shown) exhibits a sharp resonance for $E_C \approx E_X$ with an enhancement factor of ~ 25 for the vibrational energy. These results are in very good agreement with the spontaneous resonant data [38].

III. BOSE-EINSTEIN CONDENSATION AND COHERENT CONTROL OF EXCITONS

III.1 Coherent Light Emission from Exciton Condensates

A system of bosons or paired fermions is said to be collectively condensed when it exhibits correlations that reflect the macroscopic occupation of a single microscopic state. Condensation can be described in terms of a non-zero order parameter corresponding to the thermal average of an operator that creates either a boson or a pair of fermions [38-41]. In the condensed phase, the system is, locally, in a coherent superposition of eigenstates with a varying number of particles [41]. Condensation has been clearly established for superconductors and superfluid ^3He in the case of pairs of fermions and for superfluid ^4He and atomic Bose condensation in the case of bosons.

The condensation of an exciton gas in semiconductors has been extensively discussed in the literature [39, 42-56]. The transition takes place when the system meets the so-called quantum degeneracy criterion $n\lambda^D > 1$. Here, λ is the De Broglie thermal wavelength, n is the particle density and D is the number of spatial dimensions [39]. Another relevant dimensionless quantity for excitonic systems is naD where a is the exciton radius. For $naD \sim 1$ excitons behave like weakly interacting bosons, otherwise they must be treated as pairs of fermions [42,44,45]. For $naD > 1$, condensation in the exciton gas can be expected in two flavors depending on the exciton size: a genuine Bose condensation for $na^D \gg 1$ and, in the opposite limit, a BCS-like transition as for superconductors or ^3He . The finite lifetime of excitons represents an obvious drawback to achieve condensation. However, we note that typical lifetimes

are long enough for the excitons to reach a quasi thermodynamic equilibrium, which can be studied using time-resolved techniques [47].

The condensation of optically active (OA) excitons has received considerable experimental [46,47] and theoretical [42-46,49,50,52-54, 56] attention in the past several years. In such a system a crucial question arises as to the character of the light emitted by the condensate. Superradiance and coherent emission have been considered in the past [46,49,52,53,56] but, to the best of our knowledge, a definite discussion of emission properties has not been given before [51]. In our work, we considered the light emitted from a condensate of OA excitons, created using a fast laser pulse, in semiconductor Quantum Well (QW) structures. Irrespective of the process that drives the excitons to condense after the laser is turned off, we proved that: (i) the emitted light is a coherent electromagnetic field (EMF) of the Glauber type [58]; (ii) condensation of OA excitons can take place at finite temperatures in an infinite and flat two-dimensional (2D) system. This is significantly different from the case of an *isolated* 2D system for which Hohenberg theorem [59] forbids condensation at finite temperatures. The importance of (i) is twofold. First, the coherent emission could be used to probe exciton condensation. Second, the emission from the condensate is expected to occur at any density even if there is no population inversion [53]. Therefore, it constitutes a physical phenomenon different from that of lasing in high density exciton gases where coherent emission follows from population inversion [50].

III.2 Quantum Beats and Coherent Control of Excitons

The coherent response of GaAs quantum wells (QW) to ultrafast laser pulses has been the object of intense research in the last decade. More recently, the use of resonant, low-intensity subpicosecond pulses, at low temperatures, has provided a wealth of information on the exciton *linear response* [60-71]. In these "linear" experiments [72], coherence is revealed by some interference phenomena which, broadly speaking, can be classified into three groups: (i) Temporal coherent control. This refers to the case where a single excitonic resonance is driven by *two* identical laser pulses [61,64,65,67] whose mutual delay can be controlled with attosecond precision. The main feature of coherent control is that some property of the system (e.g., the exciton density [61] or optical orientation [64]) is an oscillating function of the delay. The amplitude of the oscillations goes to zero in a time scale referred to as T_2 . (ii) Resonant Rayleigh scattering (RRS). A QW excited by a laser pulse with in-plane momentum $\mathbf{k}_{||}$ emits coherent light [60,62,66,67,69-71] in non-phased-matched directions $\mathbf{q}_{||} \neq \mathbf{k}_{||}$. This is due to disorder, which breaks the translational invariance along the QW plane producing elastic scattering events which preserve the phase coherence. (iii) Light-heavy hole exciton (lx-hx) beats, which take place whenever the pulses overlap in energy with both the lx- and hx-states. Beats are observed, for instance, in the time resolved intensity of the RRS [60,62,66,67,70,71] or in time resolved reflectivity [61,62]. It must be noted that all (i), (ii) and (iii) can be observed simultaneously in the same experiment [67,68].

Interferences do take place, but *what* is interfering? There are two apparently different possibilities: (i) interferences involving the induced electric dipole and (ii) quantum interferences of the exciton wavefunction. Each possibility is related to a different kind of theoretical picture. The first one relies on the classical picture in which an

electric dipole linearly proportional to the exciting electric field of the laser is induced in the QW, considered as a dielectric [73-74]. The induced macroscopic dipole emits radiation that decays in a typical time T_2 (usually much shorter than the radiative decay time T_1). The second possibility brings us to a microscopic picture in which the laser creates a quantum coherent superposition state of excitons that decays in the time T_2 .

The primary goal of our work was to provide an unambiguous description of excitonic coherence and interferences. To this end, we used a bosonic description of excitons created by means of resonant optical pumping in a quantum-well in the presence of a weak disorder potential. We found that *excitonic coherence*, as revealed by *linear* experiments, is a many non-interacting exciton phenomenon, exactly like optical coherence is a many (non interacting) photon phenomenon [31]. In analogy with Glauber theory of photonic coherence [31], we obtained a description of excitonic coherence, in the presence of a weak disorder potential, in terms of a quantum field theory of excitons. It must be noted that our theory unifies the electromagnetic and quantum pictures mentioned earlier. In the limit of a high number of quanta, and under certain conditions, quantum fields behave classically, in the sense that their mean values are much larger than the fluctuations. In that situation electromagnetic (classical) interferences dominate. In the opposite limit (low number of quanta) the quantum behavior becomes the norm.

Disorder plays a central role in the experiments and, in particular, in time-resolved Rayleigh scattering [60,62,63,67-71]. Here, the decay of the macroscopic dipole [60-71] correlates with the finite exciton linewidth [73,75], which reflects dephasing due to the disorder. Our microscopic description is based on a second quantized Hamiltonian of non-interacting *bosonic* excitons in a weak disorder potential. The same Hamiltonian has been used in other papers in this field [76]. This model is used to address recent Rayleigh scattering experiments, by considering a quantum theory or light emission from coherent excitons. We show that RRS has a coherent

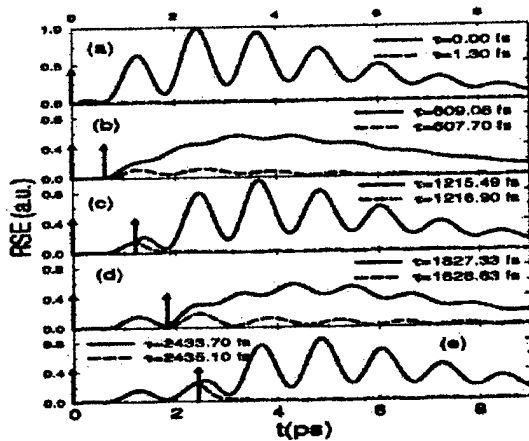


FIGURE 7. RRS for a QW excited with two pulses, the first at $t = 0$ and the second at $t = T$ with (a) $\tau \approx 0$, (b) $\tau \approx 0.6$ ps, (c) $\tau \approx 1.2$ ps (d) $\tau \approx 1.8$ ps and (e) $\tau \approx 2.4$ ps. Solid (dashed) lines represent constructive (destructive) interference. $\Gamma_h = 0.20$ meV, $\Gamma_l = 0.34$ meV.

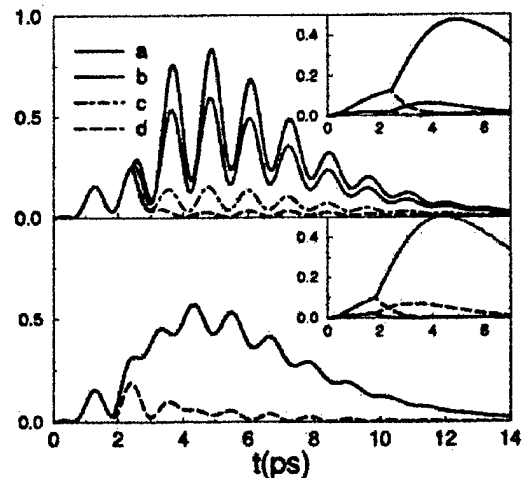


FIGURE 8. RRS in the case of enhancement (upper part) and suppression (lower part). Upper part (a) $\tau = 2433.7$ fs, (b) $\tau = 2434.25$ fs, (c) $\tau = 2434.75$ fs, and (d) $\tau = 2435.0$ fs. Inset: total hx and ix emission for (a), (b) and (c). Lower part. Solid line $\tau = 1827.3$ fs and dashed line $\tau = 1828.1$ fs. In all cases, $\Gamma_h = 0.20$ meV, $\Gamma_l = 0.34$ meV and the lx-hx splitting is 3.4 meV.

component that displays lx-hx beats. We find excellent agreement with coherent control experiments on optical orientation [64] in which the exciton spin degree of freedom is the parameter of interest, with experiments on coherent control of the Rayleigh signal [67-68] and with data on control of the exciton density [61,64,67] in a single QW. As an example, our results on RRS are reproduced in Figures 7 and 8 .

IV. REFERENCES

1. S. De Silvestri, J. G. Fujimoto, E. P. Ippen, E. B. Gamble, Jr., L. R. Williams and K. A. Nelson, *Chem. Phys. Lett.* **116**, 146 (1985).
2. T. K. Cheng, S. D. Brorson, A. S. Kazeroonian, J. S. Moodera, G. Dresselhaus, M. Dresselhaus and E. P. Ippen, *Appl. Phys. Lett.* **57**, 1004 (1990).
3. G. C. Cho, W. Kütt and H. Kurz, *Phys. Rev. Lett.* **65**, 764 (1990).
4. J. M. Chwalek, C. Uher, J. F. Whittaker, G. A. Mourou, J. Agostinelly and M. Lelental, *Appl. Phys. Lett.* **57**, 1696 (1990).
5. H. J. Zeiger, J. Vidal, T. K. Chen, E. P. Ippen, G. Dresselhaus and M. S. Dresselhaus, *Phys. Rev. B* **45**, 768 (1992).
6. W. Albrecht, Th. Kruse and H. Kurz, *Phys. Rev. Lett.* **69**, 1451 (1992).
7. T. Mishina, Y. Masumoto, B. Fluegel, K. Meissner and N. Peyghambarian, *Phys. Rev. B* **46**, 4229 (1992).
8. T. Pfeifer, W. Kütt, H. Kurz and R. Scholz, *Phys. Rev. Lett.* **69**, 3248 (1992).
9. T. K. Cheng, L. H. Acioli, J. Vidal, H. J. Zeiger, G. Dresselhaus, M. S. Dresselhaus and E. P. Ippen, *Appl. Phys. Lett.* **62**, 1901 (1993).
10. T. Pfeifer, T. Dekorsy, W. Kütt and H. Kurz, *Appl. Phys. A* **55**, 482 (1992).
11. P. Basséras, S. M. Gracewski, G. W. Wicks and R. J. D. Miller, *J. Appl. Physics* **75**, 2761 (1994); A. Yamamoto, T. Mishina, Y. Masumoto and M. Nakayama, *Phys. Rev. Lett.* **73**, 740 (1994).
12. Y. Liu, A. Frenkel, G. A. Garrett, J. F. Whitaker, S. Fahy, C. Uher and R. Merlin, *Phys. Rev. Lett.* **75**, 334 (1995).
13. S. Hunsche, K. Wienecke, T. Dekorsy and H. Kurz, *Phys. Rev. Lett.* **75**, 1815.
14. T. Dekorsy, H. Auer, C. Waschke, H. J. Bakker, H. G. Roskos, H. Kurz, V. Wagner and P. Grosse, *Phys. Rev. Lett.* **74**, 738 (1995).
15. A. L. Dobryakov, V. S. Letokhov, Yu. E. Lozovik, Yu. A. Matveets, V. M. Farztdinov, S. A. Kovalenko, N. R. Ernsting and G. Marovskii, *JETP Lett.* **61**, 985 (1995).
16. T. Dekorsy, A. M. T. Kim, G. C. Cho, H. Kurz, A. V. Kutznetsov and A. Förster, *Phys. Rev. B* **53**, 1531 (1996).
17. D. P. Kien, J. C. Loulergue and J. Etchepare, *Phys. Rev. B* **47**, 11027 (1993).
18. H. J. Bakker, S. Hunsche and H. Kurz, *Phys. Rev. B* **50**, 914 (1994).
19. T. P. Dougherty, G. P. Wiederrecht, K. A. Nelson, M. H. Garrett, H. P. Jensen and C. Warde, *Science* **258**, 770 (1992)..
20. P. Grenier, D. Houde, S. Jandl and L. A. Boatner, *Phys. Rev. B* **47**, 1 (1993).
21. G. P. Wiederrecht, T. P. Dougherty, L. Dhar, K. A. Nelson, D. E. Leaird and A. Weiner, *Phys. Rev. B* **51**, 916 (1995).
22. J. L. Macho, F. W. Wise, R. C. Patel and D. B. Tanner, *Phys. Rev. B* **48**, 2819 (1993).
23. G. A. Garrett, T. F. Albrecht, J. F. Whitaker and R. Merlin, *Phys. Rev. Lett.* **77**, 3661 (1996).
24. M. Hase, K. Mizoguchi, H. Harima, S. Nakashima, M. Tani, K. Sakai and M. Hangyo, *Appl. Phys. Lett.* **69**, 2474 (1996).
25. Y. -X. Yan, E. B. Gamble, Jr. and K. A. Nelson, *J. Chem. Phys.* **83**, 5391 (1985); Y. -X. Yan and K. A. Nelson, *ibid.* **87**, 6240 (1987) and *ibid.* **87**, 6257 (1987).
26. S. Ruhman, B. Kohler, A. G. Joly and K. A. Nelson, *IEEE J. Quantum Electron.* **24**, 460 (1988).
27. W. A. Kütt, W. Albrecht and H. Kurz, *IEEE J. Quantum Electron.* **28**, 2434 (1992).
28. L. Dhar, J. A. Rogers and K. A. Nelson, *Chem. Rev.* **94**, 157 (1994).
29. R. Merlin, *Solid State Commun.* **102**, 207-220 (1997).
30. G. A. Garrett, A. G. Rojo, A. K. Sood, J. F. Whitaker and R. Merlin, *Science* **275**, 1638 (1997).

31. R. J. Glauber, *Phys. Lett.* **21**, 650 (1966).
32. See, e.g., C. Cohen-Tannoudji, J. Dupont-Roc and G. Grynberg, *Atom-Photon Interactions - Basic Processes and Applications* (Wiley, N. Y. 1992); D. F. Walls and G. J. Milburn, *Quantum Optics* (Springer, Berlin, 1994).
33. T. E. Stevens, J. Kuhl and R. Merlin, *Phys. Status Solidi (b)* **215**, 81-86 (1999); presented at the International Conference on Solid State Spectroscopy, 1999, Schwäbisch Gmünd, Germany.
34. G. A. Garrett, J. F. Whitaker and R. Merlin, in *Ultrafast Phenomena XI*, ed. by T. Elsaesser, J. G. Fujimoto, D. A. Wiersma and W. Zinth (Springer, Berlin, 1998), p.362.
35. See, e.g., R. Blinc, *Soft Modes in Ferroelectric and Antiferroelectrics*, (North-Holland, Amsterdam, 1974); *Light Scattering near Phase Transitions*, ed. by H. Z. Cummins and A. P. Levanyuk (North-Holland, Amsterdam, 1983), and references therein.
36. P. A. Fleury and J. M. Worlock, *Phys. Rev.* **174**, 613 (1968).
37. K. A. Müller, W. Berlinger and E. Tosatti, *Z. Phys. B* **84**, 277 (1991); D. E. Grupp and A. M. Goldman, *Science* **276**, 392 (1997).
38. R. M. Hoff and J. C. Irwin, *Phys. Rev. B* **10**, 3464 (1974); J. Reydellet and J. M. Besson, *Solid State Commun.* **17**, 23 (1975).
39. *Bose Einstein Condensation*, (Cambridge University Press, Cambridge, 1995); ed. by A. Griffin, D. W. Snoke and S. Stringari.
40. D. Forster, *Hydrodynamic Fluctuations, Broken Symmetry and Correlation Functions* (Benjamin, New York, 1975).
41. P. W. Anderson, *Basic Notions of Condensed Matter Physics* (Benjamin, Menlo Park, 1984).
42. L. V. Keldysh and Yu. V. Kopayev, *Fiz. Tverd. Tela*, **6**, 2791 (1964) [*Sov. Phys. Solid State* **6**, 2219 (1965)]; D. Jerome, T. M. Rice and W. Kohn, *Phys. Rev.* **158**; E. Hanamura and H. Haug, *Phys. Rep.* **33**, 209 (1977).
43. V. A. Gergel *et al.*, *Zh. Eksp. Teor. Fiz.* **53**, 544 (1967) [*Sov. Phys. JETP*, **26**, 354 (1968)].
44. C. Comte and P. Nozieres, *P., J. Physique* **43** 1069 (1982); P. Nozieres and C. Comte, *ibid.* **43** 1083 (1982).
45. H. Haug and S. Schmitt-Rink, *Prog. Quant. Electr.* **9**, 3 (1984).
46. T. Fukuzawa *et al.*, *Phys. Rev. Lett.* **64**, 3066 (1990); *Surface Sci.* **228**, 482 (1990); J. Kash *et al.*, *Phys. Rev. Lett.* **66**, 2247(1991).
47. L. V. Butov *et al.*, *Phys. Rev. Lett.* **73**, 304 (1994).
48. J. P. Wolfe *et al.* in [39].
49. X. Zhu *et al.*, *Phys. Rev. Lett.* **74**, 1633 (1995); P. B. Littlewood and X. Zhu, *Physica Scripta*, **T68**, 56 (1996).
50. M. E. Flatte *et al.*, *Appl. Phys. Lett.* **66**,1313 (1995).
51. See, e.g. D. W. Snoke, *Science* **273**, 1351 (1996) and references therein; S. A. Moskalenko and D. W. Snake, *Bose-Einstein Condensation of Excitons and Biexcitons*; Cambridge University Press, to be published.
52. Th. Oestreich *et al.*, *Solid State Commun.* **100**, 325 (1996).
53. A. Imamoglu *et al.*, *Phys. Rev. A* **53**, 4250 (1996).
54. J. Fernandez-Rossier and C. Tejedor, *Phys. Rev. Lett.* **78**,4809 (1997).
55. S. Conti *et al.*, *Phys. Rev. B* **57**, R6864 (1998).
56. B. Laikhtman, *Europhys. Lett.*, to be published.
57. J. Shah, *Hot Carriers in Semiconductor Nanostructures* (Academic Press, San Diego, 1992).
58. R. J. Glauber, *Phys. Rev.* **131**, 2766 (1963).
59. P. C. Hohemberg, *Phys. Rev.* **158**, 383 (1967).
60. H. Wang *et al.*, *Phys. Rev. Lett.* **14**, 3065 (1995).
61. A. P. Heberle, J. J. Baumberg, K. Koehler, *Phys. Rev. Lett.* **15**, 2598 (1995); J. J. Baumberg *et al.*, *Phys. Stat. Sol (b)* **204**, 9 (1997).
62. S. Haacke *et al.*, *Phys. Rev. Lett.* **18**, 2228 (1997); in *Trends. in Optics and Photonics* **18**, ed. by D. Citrin (Optical Society of America, 1998).
63. M. Gurioli *et al.*, *Phys. Rev. Lett.* **18**, 3205 (1997); Ceccherini *et al.*, *Optics Commun.* **132**, 77 (1996)
64. X. Marie *et al.*, *Phys. Rev. Lett.* **78**, 3222 (1997); P. Le Jeune *et al.*, *Phys. Stat. Sol. (a)* **184**, 527 (1997).
65. J. J. Baumberg *et al.*, *Phys. Rev. Lett.* **80**, 3567 (1998).
66. D. Birkedal and J. Shah, *Phys. Rev. Lett.* **81**, 2372 (1998).
67. M. Wörner and J. Shah, *Phys. Rev. Lett.* **81**, 4208 (1998).
68. S. P. Kennedy *et al.*, in *Proceedings of ICPS 24*, ed. by D. Gershoni (World Scientific, Singapore, 1998), to be published; N. Garro *et al.*, *Phys. Rev. B* **80**, 4497 (1999).
69. W. Langbein, J. Hvam and R. Zimmermann, *Phys. Rev. Lett.* **82**,1040 (1999).

70. S. Haacke *et al.*, preprint.
71. A. V. Shchegrov, D. Birkedal, J. Shah, *Phys. Rev. Lett.* **83**, 1391 (1999)
72. The term "linear" refers to experiments for which the dominant features of the interference originate from linear response and the exciton density is low.
73. H. Stolz, *Time-Resolved Light Scattering from Excitons* (Springer, Berlin, 1994).
74. I. G. Lang, V. I. Belitsky and M. Cardona, *Phys. Stat. Sol. (a)* **184**, 307 (1997).
75. J. Shah, *Ultrafast Spectroscopy of Semiconductors and Semiconductor Nanostructures* (Springer, Berlin, 1996).
76. R. Zimmermann, *Nuov. Cim.* **17D**, 1801 (1995).

V. PUBLICATIONS UNDER ARO SPONSORSHIP

V.1 JOURNALS

1. "Coherent THz phonons driven by light pulses and the Sb-problem: what is the mechanism?" G. A. Garrett, T. F. Albrecht, J. F. Whitaker, and R. Merlin, *Phys. Rev. Lett.* **77**, 3661-3664 (1996).
2. "Generating coherent THz phonons with light pulses." R. Merlin, in *Highlights of Condensed Matter Physics*, *Solid State Commun.* **102**, 207-220 (1997).
3. "Vacuum-squeezing of solids: macroscopic quantum states driven by light pulses." G. A. Garrett, A. G. Rojo, A. K. Sood, J. F. Whitaker and R. Merlin, *Science* **275**, 1638-1640 (1997).
4. "Ultrafast optical excitation of a combined coherent-squeezed phonon field in SrTiO₃." G. A. Garrett, J. F. Whitaker, A. K. Sood and R. Merlin, *Optics Express* **1**, 385-389 (1997).
5. "Coherent Light Emission from Exciton Condensates in Semiconductor Quantum Wells." J. Fernandez-Rossier, C. Tejedor and R. Merlin, *Solid State Commun.* **108**, 473-477 (1998).
6. "Stress Effects in GaAs: Anomalous Selection Rules and Heavy-Light Hole Beats." N. H. Bonadeo, D. G. Steel and R. Merlin, *Phys. Rev. B.* **60**, 8970-8974 (1999).
7. "The Phonon Bragg Switch: A Proposal to Generate Sub-Picosecond X-Ray Pulses." P. H. Bucksbaum and R. Merlin, *Solid State Commun.* **111**, 535-539 (1999).
8. "Exciton Beats in GaAs Quantum Wells: Bosonic Representation and Collective Effects." J. Fernandez-Rossier, C. Tejedor and R. Merlin, *Solid State Commun.* **112**, 597-600 (1999).

V.2 CONFERENCE PROCEEDINGS

1. "Electric field domains, Pockels effect and coherent acoustic phonons in superlattices." in *Optical Spectroscopy of Low-Dimensional Semiconductors*, ed. by G. Abstreiter *et al.* (Kluwer Academic Publishers, Dordrecht, 1997), pp. 83-97; invited paper at the NATO Advanced Study Institute on Optical Spectroscopy of Low-Dimensional Semiconductors, 1996, Ankara, Turkey.
2. "Generation of Coherent and Squeezed Phonon Fields using Femtosecond Laser Pulses." G. A. Garrett, J. F. Whitaker and R. Merlin, in *Ultrafast Phenomena XI*, ed. by T. Elsaesser, J. G. Fujimoto, D. A. Wiersma and W. Zinth (Springer, Berlin, 1998), pp.362-364; presented at the XI International Conference on Ultrafast Phenomena, 1998, Garmisch-Partenkirchen, Germany.
3. "Coherent Light Emission from Condensed Radiating Excitons." J. Fernandez-Rossier, C. Tejedor and R. Merlin, in *The Physics of Semiconductors*, Vol. , ed. by D. Gershoni (World Scientific, Singapore, 1999), pp. ; presented at the 24th International Conference on the Physics of Semiconductors, 1998, Jerusalem, Israel.
4. "Interferences and Coherent Control of Excitons in GaAs Quantum Wells." J. Fernandez-Rossier, C. Tejedor and R. Merlin, *J. Phys.: Condens. Matter* **11**, 6013-6021 (1999); invited paper at the International Workshop on Nanophysics and Electronics (NPE-98), 1998, Lecce, Italy.

V.3 THESES

1. "Ultrafast Dynamics of Low-Energy Elementary Excitations in Semiconductors." T. E. Stevens, Ph. D. Thesis, University of Michigan, 2000.

V.4 OTHER CONTRIBUTIONS

1. "The sound of a quantum loudspeaker." R. Merlin, *Physics World* **10** (8), 25-26 (1997).
2. "Squeezed states: controlling quantum noise in a solid." G. A. Garrett and R. Merlin, *LEOS Newsletter* **11** (5), 7-9 (1997).
3. "Raman Scattering in Materials Science." Springer Series in Materials Science **42** (Springer, Berlin, 2000). Edited by W. H. Weber and R. Merlin.

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