APPLIED PHYSICS LETTERS VOLUME 75, NUMBER 9 30 AUGUST 1999

Cooling of a semiconductor by luminescence up-conversion

E. Finkeißen, M. Potemski,[a] and P. Wyder
Grenoble High Magnetic Field Laboratory, MPI/FKF and CNRS,
25, avenue des Martyrs, BP166, F-38042 Grenoble, Cedex 9, France

L. Viña
Departamento de Física de Materiales, C-IV, Universidad Autónoma de Madrid,
Cantoblanco, E-28049, Madrid, Spain

G. Weimann
Walter-Schottky-Institut, Technische Universität München, D-8046 Garching, Germany

(Received 27 April 1999; accepted for publication 6 July 1999)

We report the observation of phonon-mediated up-conversion of luminescence in a GaAs quantum well. This opens the possibility of light-induced lattice cooling in a semiconductor. Under appropriate conditions, pumping the sample with light at the energy of the heavy-hole exciton, we observe light-hole exciton emission, which lies ~10 meV above the excitation energy. The use of an external magnetic field together with the resolution of excited excitonic states provides an internal thermometer to monitor the sample temperature. Temperature drops as large as 10% of the initial temperature are observed for pump densities of 4 W cm^{-2}. *© 1999 American Institute of Physics.* [S0003-6951(99)00435-0]

It has been long recognized that interaction of a system with radiation can lead to cooling.[1] In the case of atoms, Doppler cooling[2–4] and spontaneous anti-Stokes scattering[5] have been shown to be powerful techniques, which have allowed the observation of an atomic Bose–Einstein condensation.[6] Most difficult to achieve has proven to be cooling in the condense phase, although the basic thermodynamic ideas were established by Landau fifty years ago,[7] and the first experimental attempt was tried at the end of the sixties.[8] Laser cooling of a solid might be possible when resonantly pumping a given electronic state and releasing the heat from the lattice subsystem via an up-converted emission arising from higher energy electronic states excited in phonon absorption processes. Only lately two groups have reported on successful cooling in rare-earth doped glasses and a fluid solution of a laser dye.[9–12] Using laser light one may expect the possibility of local cooling effects, which are of particular importance in case of semiconductor structures.

Here we present direct evidence of local cooling effects in a representative semiconductor structure with temperature drops of the order of 7 K from liquid nitrogen temperature. A two dimensional semiconductor—intrinsic GaAs/$\mathrm{Ga}_{1-x}\mathrm{Al}_x\mathrm{As}$ quantum well (QW)—exhibits certain peculiarities, which make it an ideal system to study cooling. First, samples with high quantum efficiency are obtained by epitaxial growth. Second, the spectra of QWs are dominated by excitonic effects, which implies sharp, nonoverlapping lines: the energy separation of heavy- (hh) and light-hole (lh) excitons can be tailored by choosing the appropriate well thickness and Al content. Finally, an external magnetic field supplies an internal thermometer: The field favors the observation of excited excitonic states.[13] The simultaneous presence of the up-converted PL from lh$(1s)$ and the first excited hh exciton, hh$(2s)$, allows us to fit their intensities using a thermalized Boltzmann distribution, without the need of any external device.

The structure we selected consists of three uncoupled 90-Å-wide GaAs QWs with 0.1 μm $\mathrm{Ga}_{0.74}\mathrm{Al}_{0.26}\mathrm{As}$ barriers grown on semi-insulating GaAs. The sample has a high quantum efficiency: Only a very weak temperature dependence of the hh$(1s)$-PL intensity is observed in the range of 2–60 K. The sample, mounted in a variable temperature cryostat, was excited with a Ti:sapphire laser. Magnetic fields up to 16 T were applied in the Faraday configuration.

As illustrated in Fig. 1, photoluminescence excitation (PLE) spectra show two main peaks at $B=0$ which correspond to hh$(1s)$ and lh$(1s)$. New peaks appear when a magnetic field is applied. The spectra have been measured in the configuration of σ^- circularly polarized emission and excitation light. The strongest features correspond to the ''s'' states.[13] We will concentrate on the peaks hh$(1s)$, lh$(1s)$, and hh$(2s)$, whose field dependence is shown in the inset. The appearance of three clear excitonic resonances is essential for our method of monitoring the local temperature.

The proof for anti-Stokes PL is illustrated in Fig. 2. PLE and PL ($T=50$ K, $B=7$ T) are shown in Figs. 2(a) and 2(b), respectively. Exciting at 1.579 eV, three peaks are observed in the PL (b) with a direct correspondence to those in the PLE (a). The thermal equilibrium of the carriers with the lattice populates the higher states, which therefore are observed in emission. The power density was 5 mW cm^{-2}, low enough to avoid heating. Thus, knowing the lattice T, one can obtain the ratio of oscillator strengths (f) of lh$(1s)$ and hh$(2s)$ from their intensities (I): $\alpha = f[\mathrm{hh}(2s)]/f[\mathrm{lh}(1s)] = I[\mathrm{hh}(2s)]/I[\mathrm{lh}(1s)] \times e^{\Delta/kT}$, where Δ is the energy difference $E[\mathrm{hh}(2s)]-E[\mathrm{lh}(2s)]$, and k is the Boltzmann constant. The intensities of lh$(1s)$ and hh$(2s)$ assuming a Boltzmann distribution for $\alpha=0.81$ and $T=50$ K are shown as open circles. The value obtained for α is in agreement with calculations of magnetoexcitons in GaAs QWs.[13]

---

[a] Electronic mail: potemski@labs.polycnrs-gre.fr

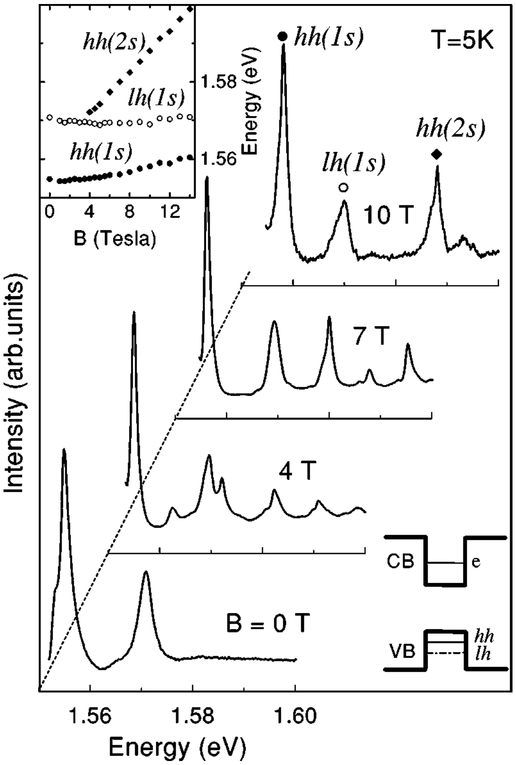

FIG. 1. Low temperature photoluminescence excitation spectra of the 90-Å-wide GaAs QWs at different magnetic fields. The inset shows the field dependence of the energies corresponding to the ground state excitions, hh(1s), lh(1s), and the first excited state hh(2s).

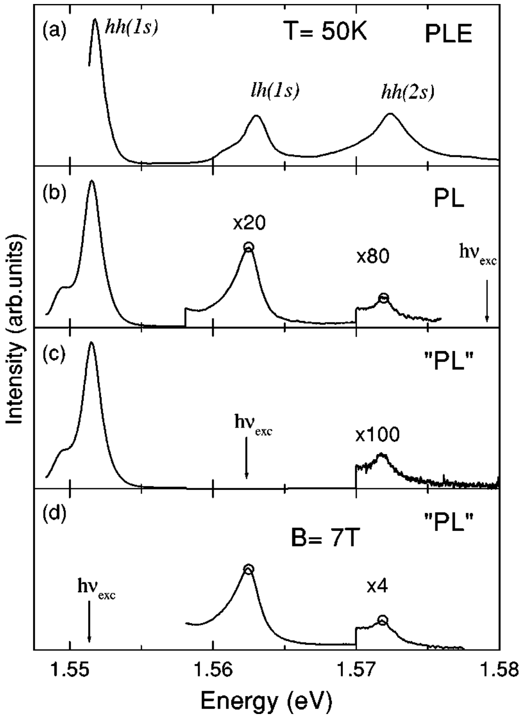

FIG. 2. Comparison of photoluminescence excitation (a) and normal photoluminescence (b) at 50 K for a magnetic field strength of 7 T. Panels (c) and (d): photoluminescence for different excitation energies marked by the arrows. Some regions of the spectra are magnified by the factors shown in the figure.

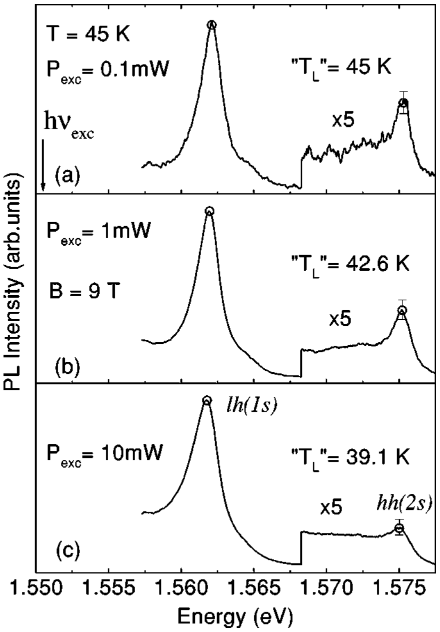

FIG. 3. Up-converted luminescence at a bath temperature of 45 K in a magnetic field of 9 T for three different excitation powers: (a) 0.1, (b) 1, and (c) 10 mW. The excitation energy was at the hh(1s), marked by the arrow. Circles: intensities deduced from a Maxwell distribution yielding a temperature T_L (error bars correspond to ±1.5 K).

More conspicuous is the finding of Fig. 2(c): Exciting at lh(1s) with $5\,\mathrm{mW\,cm^{-2}}$, we observe the emission of hh(1s), and also an up-converted signal from hh(2s). If the sample is excited at hh(1s), we observe anti-Stokes emission from both lh(1s) and hh(2s) [Fig. 2(d)]. The presence of both peaks allows an estimation of the lattice temperature. The open circles are again obtained with $\alpha=0.81$ and $T=50\,\mathrm{K}$. We found that at the low excitation power, the intensity ratio between lh(1s) and hh(2s) emission peaks is independent whether the excitation energy is high and the ''usual'' PL is detected (''hot'' carriers thermalize with a ''cold'' lattice) or the excitation energy is resonant with the hh(1s) exciton and the up-converted emission is observed (cold carriers thermalize with a ''warm'' lattice).[14]

Figure 3 demonstrates the cooling effect through the establishment of an equilibrium among hh(1s) and lh(1s) $-$hh(2s). Having established that low illumination powers do not alter the temperature (Fig. 2), we adjusted α in Fig. 3(a) to recover a lattice temperature, ''T_L,'' equal to the bath temperature, $T=45\,\mathrm{K}$. The circles depict the intensities expected from a Boltzmann distribution with temperature T_L (error bars ±1.5 K). Increasing the excitation power [Fig. 3(b)], the ratio of lh(1s) and hh(2s) peaks changes notably: Keeping the same α, $T_L=42.6\,\mathrm{K}$ is obtained. A further power increase [Fig. 3(c)] gives a T_L drop of 5.9 K. In our experiments, lattice cooling takes place under continuous excitation and only within the area of the laser spot. This implies that a strongly coupled electronic-phonon system exists in this spot, which must be somehow disconnected from the nonilluminated part of the sample. Since the coupling between these two subsystems cannot be easily assessed, our results must be considered only as qualitative.

A closer examination of lh(1s) in Fig. 3(c) (laser power 10 mW) reveals a slight broadening at high powers. In order to eliminate the complex behavior of the exciton gas, which might be expected at higher densities, in the following we concentrate on experimental results obtained in the range of low excitation powers ($P\leq 8\,\mathrm{mW}$). In this range, the shape

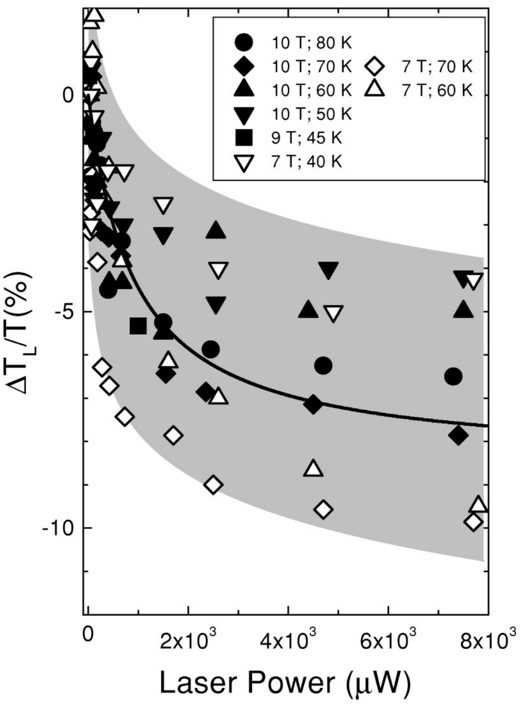

FIG. 4. Temperature drop, in percentage, as a function of excitation power for different bath temperatures (T) and magnetic fields (symbols). The line depicts the results of a modeling described in the text.

of the measured spectra is not sensitive to the excitation power. A compilation of our results is presented in Fig. 4, which shows the percentage of temperature decrease as a function of P. In spite of the large data spread, a clear tendency of cooling is obtained, showing saturation at high powers. The line represents the results of a qualitative modeling of the energy exchange between an electronic and a phonon system. It considers thermal equilibrium, under continuous-wave (cw) excitation, among two electronic levels, separated by an energy Δ, and the population of phonons

$$n_{\mathrm{ph}}=\frac{1}{e^{\Delta/kT}-1},$$

simulated by a phonon of energy $\hbar\omega=\Delta$. Only the lowest level is populated by illumination and a ratio of radiative lifetimes $\tau[\mathrm{up}]/\tau[\mathrm{down}]=3$ is assumed.[15] Heat is extracted from the lattice through emission from the upper level. The energy transfer from the rest of the sample to the illuminated spot is mimicked by an increase of n_{ph} proportional to the difference between $n_{\mathrm{ph}}(T)$ and $n_{\mathrm{ph}}(T_L)$. It is also supposed that $\sim$5% of P heats the sample. This model, in spite of its naiveté, reproduces well the observed behavior and explains the saturation of the cooling, which has been suggested[11] but not measured before.

In summary, we have demonstrated anti-Stokes emission in semiconductor QWs. Our results might be stimulating for alternative studies of carrier–phonon interactions (phonon absorption processes), when, for example, applying time-resolved techniques to investigate the up-converted emission. The possibility of local cooling has been qualitatively shown. The application of an external magnetic field allows a non-intrusive determination of the temperature in a simple experiment without requiring any sophisticated conditions. By using recent advances in optoelectronic integration, the local cooling of electronic devices by up-conversion pumping with an efficient diode laser could be conceivable.

L.V. acknowledges the Joseph-Fourier University (Grenoble) for its hospitality. Part of this work has been performed under the ‘‘Picasso’’ collaboration program.

[1] P. Z. Pringsheim, Phys. **57**, 739 (1929).
[2] T. W. Hansch and A. L. Schawlow, Opt. Commun. **13**, 68 (1975).
[3] C. N. Cohen-Tannoudji and W. D. Philips, Phys. Today **43**, 33 (1990).
[4] S. Chu, Science **253**, 861 (1991).
[5] N. Djeu and W. T. Whitney, Phys. Rev. Lett. **46**, 236 (1981).
[6] M. H. Anderson, J. R. Ensher, M. R. Matthews, C. E. Wieman, and E. A. Cornell, Science **269**, 198 (1995).
[7] L. Landau, J. Phys. (Moscow) **10**, 503 (1946).
[8] T. Kushida and J. E. Geusic, Phys. Rev. Lett. **21**, 1172 (1968).
[9] R. I. Epstein, M. I. Buchwald, B. C. Edwards, T. R. Gosnell, and C. E. Mungan, Nature (London) **377**, 500 (1995).
[10] C. E. Mungan, M. I. Buchwald, B. C. Edwards, R. I. Epstein, and T. R. Gosnell, Phys. Rev. Lett. **78**, 1030 (1997).
[11] J. L. Clark and G. Rumbles, Phys. Rev. Lett. **76**, 2037 (1996).
[12] A. Kastler, J. Phys. Radium **11**, 255 (1950).
[13] See, e.g., Y. Iimura, Y. Segawa, G. E. W. Bauer, M. M. Lin, Y. Aoyagi, and S. Namba, Phys. Rev. B **42**, 1478 (1990); M. Potemski, L. Viña, J. C. Maan, G. E. W. Bauer, K. Ploog, and G. Weimann, *ibid.* **43**, 14707 (1991), and references therein.
[14] See, for example, E. O. Goebel and O. Hilderband, Phys. Status Solidi B **88**, 645 (1978).
[15] Corresponding to $\tau[\mathrm{lh}(1s)]/\tau[\mathrm{hh}(1s)]$.