Resonant Rayleigh Scattering in Ordered and Intentionally Disordered Semiconductor Superlattices

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We report the experimental study of resonant Rayleigh scattering in GaAs-AlGaAs superlattices with ordered and intentionally disordered potential profiles (correlated and uncorrelated) in the growth direction $z$. We show that the intentional disorder along $z$ modify markedly the energy dispersion of the dephasing rates of the excitons. The application of an external magnetic field in the same direction allows the continuous tuning of the in plane exciton localization and to study the interplay between the in plane and vertical disorder.

INTRODUCTION

In recent years, a large number of studies has been devoted to explore the general properties of disordered systems through the investigation of the optical response and transport properties of semiconductor nanostructures. An important topic in the physics of disordered systems is to understand the influence of an intentionally disordered potential profile along the growth axis $z$ in semiconductor superlattices (SL’s) on quantum interference and carrier localization [1, 2, 3]. SL’s are excellent candidates to perform such a study since the advances achieved in molecular beam epitaxy (MBE) allow the fabrication SL’s tailored with desired conduction- and valence-band profiles. In this way it is possible to design a great variety of one dimensional potentials with intentional disordered in the growth direction $z$ (vertical disorder) which coexists with the in plane disorder due to unintentional growth islands. The global result is an anisotropic disorder which effect on the quantum interference has become an interesting subject of recent experimental studies. As an example, magneto-transport measurements in doped semiconductor superlattices recently allowed the study of the effects of the anisotropy on the phase-breaking time of the electrons [2].

In the present work we measure simultaneously the continuous wave resonant Rayleigh scattering (RRS), the photoluminescence(PL) and photoluminescence excitation (PLE) spectra of semiconductor SL’s in a magnetic field to study the influence of the vertically correlated and uncorrelated anisotropic disorder on the coherence relaxation and exciton localization. We use this technique since RRS has access to information on the effective localization and on the spectral dispersion of the dephasing [4] while the photoluminescence(PL) and photoluminescence excitation (PLE) spectra in magnetic field are valid techniques to study the exciton delocalization along $z$ [5]. At the same time the external magnetic field allows to continuously tune the in plane exciton localization, with respect to the vertical one.

RRS is the resonant light scattering into all directions due to the localization of the exciton center of mass wave functions, and is proved to be a powerful tool for the study of dephasing process mainly in quantum wells (QWs) [6, 7, 8, 9, 10, 11, 12, 13, 14, 15], but also in microcavities [16], quantum wires [17] and in bulk semiconductors [11, 15]. Rayleigh scattering in semiconductors arises form imperfection breaking the symmetry of translational invariance of the crystal due to imperfection in the bulk or on the surface, or from interfaces. However crystals of very high quality can be grown by means of MBE, and in this crystals the defect scattering can be extremely low. In such systems and in particularly in GaAs-AlGaAs multiple QWs, Hegarty and coworkers for the first time observed a strong contribution to the Rayleigh scattering in the vicinity of an optical resonance [6, 7]. In their pioneering experiment the authors observed that RRS was enhanced 200-fold with respect to the non resonant surface and defect scattering. This RRS arises from the spatial fluctuation in the local resonant frequency which, because of the strong dispersion near resonance, leads to corresponding fluctuations in the refractive index. Hegarty et al. showed also that RRS scattering is a valid experimental technique to study the localization/delocalization of two dimensional excitons in QWs [7]. Despite the analogy between photoluminescence (PL) and RRS, these two phenomena have a
completely different natures. PL is an incoherent process due to the relaxation of a real exciton population. However, RRS derives from the coherent scattering of light as a consequence of the relaxation of wave-vector in a real solid and mainly originate from localized exciton states [8, 11, 14].

Experimental spectroscopic techniques like time resolved RRS and RRS spectral speckle analysis have been recently used with success in order to extract quantitative information on structural disorder from RRS (see, for instance Ref. 4 and references therein). In particular, the measured spectrally and time resolved RRS spectra have been analyzed with accurate models which calculate the RRS taking into account all the important features of semiconductor heterointerface fabrication. Specifically Savona and coworkers used either a time propagation of the excitonic polarization with a phenomenological dephasing or a full excitonic eigenstate model including microscopically calculated dephasing rates and phonon scattering rates [3]. The experimental spectra well agree with the calculations demonstrating the role of many elements of the disorder affect the RRS [4, 19]. Between these elements we have the interface structure, correlations and anisotropy.

EXPERIMENTAL

In this letter we study the RRS, PL and PLE in Al0.35Ga0.65As/GaAs ordered, intentionally disordered and correlated disordered SL in an external magnetic field up to 14 T. The excitation light was supplied by a Ti-sapphire laser pumped by an Ar+ ion laser. The excitation laser light was polarized \( \sigma^+ \) and the component \( \sigma^- \) of the scattered and emitted light has been detected. The samples are three \( n^+-i-n^+ \) heterostructures with a SL grown in the intrinsic region, grown by MBE. All the three samples have a SL of 200 periods and Al0.35Ga0.65As barriers with width of 3.2 nm. In the ordered SL (OSL) all the 200 wells are identical with well width 3.2 nm (hereafter referred to as A wells). In the random SL (RSL), it contains 142 A wells and 58 wells of thickness 2.6 nm (hereafter referred to as B wells) and this B wells appears randomly. The so-called random dimer SL (DSL) is identical to the RSL with the additional constraint that the B wells appear only in pairs. In the latter sample the disorder exhibit the desired short-range spatial correlations. Additional details on the samples can be found in previous publications [8, 20].

Our previous theoretical and experimental results [3, 20, 21] showed that whereas in the RSL the states are localized, the DSL supports a narrow band of critical (non-Bloch like) extended states, while the OSL supports a wide band of Bloch extended states. Therefore in the OSL the carrier are mobiles either in the growth direction and in the plane. Instead for RSL and DSL we found that the mobility is intimately related to the disorder and that the exciton dynamics is affected by the presence of correlations that completely modify the localization of the electronic states in the growth direction [3, 21].

RESONANT RAYLEIGH SCATTERING AND PHOTOLUMINESCENCE EXCITATION

In Fig. 1 we report a series of emission spectra (luminescence plus scattered light) measures on the OSL (a), DSL (b) and RSL (c) respectively, scanning the energy of the excitation laser light, for several hundreds different values of the excitation energy for polarized excitation and detection. These emission spectra have been plotted using small dots, and two main structures can be identified, one with high density of points corresponding to PL and the other with less dense point corresponding to RRS. For clarity, in the insert of the upper panel of Fig. 1(a) we show only 19 of these spectra (for which the excitation energy differs of 0.8 meV from one spectrum to the other). When the laser energy is set off resonance we observe a weak Rayleigh scattering due to defect and imperfections. Instead when the laser energy is set close to the resonance the Rayleigh scattering increases dramatically and reaches a peak some tenths times the off-resonance defect scattering. As far as we know this is the first report of RRS in semiconductor superlattices. In the same figure we include the photoluminescence excitation spectra (PLE) measured detecting at the energy of the maximum of the PL peak. The PLE spectrum of the OSL (Fig. 1(a)) shows a main peak due to the heavy hole exciton (HH) and a smaller one due to the light hole one (LH). This attribution of the LH transition is in agreement with the recent work of Gerlovin et al. who measured PLE spectra in SL samples with similar well and barrier widths [22]. As regards the DSL, its PLE spectrum (Fig. 1(b)) shows of a main peak at 1.698 eV due to the transition between the valence and conduction minibands which are present due to the dimer type correlation of the disorder given by the paired 2.6 nm wells. The spectrum present also a wider structure at 1.707 eV which is due to excitonic transitions in the 3.2 nm isolated wells. These two structure are observed also in the PL spectrum and in the RRS since the latter is a scattering phenomenon resonant with the PL. Finally for the RSL (Fig. 1(c)) the PLE and the RRS spectrum of this sample at zero magnetic field shows a main structure with a high energy exponential tail typical of the disordered systems.

As regards the analysis of the RRS, Hegarty et al. [6] in their pioneering experimental study in semiconductor QWs interpreted the experiments with the presence of a mobility edge to explain the dispersion of the dephasing time. Their work was described theoretically by Takagahara who studied in detail the link between the energy
dependence of the homogeneous linewidth and the dispersion of the dephasing time \( \Gamma_h \). More recent theories and experiments show that the RRS can be interpreted without the need of introducing a mobility edge [4, 24]. In particular Savona et al. developed very accurate theoretical calculations of the RRS using exciton eigenstates and microscopically calculated dephasing rates due to radiative decay and phonon scattering, which are in very good agreement with the experiments [4].

The observation of a shift between RRS and PLE in semiconductor QWs for the HH transition was interpreted as a consequence of the inhomogeneous broadening of the excitonic transition [6, 7, 8, 9, 10, 11]. In contrast in bulk GaAs and in GaAs-AlGaAs QWs intentionally grown with high amount of interfacial disorder, the shift of the RRS with respect to the PLE was found to be negligible [11]. Accordingly to the description of Hegarty et al. [6] the shift is due to a rapid increase of the homogeneous linewidth \( \Gamma_h \). Instead the absence of a shift between RRS and PLE can be interpreted as a constant value for \( \Gamma_h \) either in the case of localized or propagating excitons. With respect to the origin of the shift between RRS and PLE in semiconductor QW’s some authors ascribed it to the presence of a mobility edge between propagating and localized states [6, 7, 9]. However, as we mentioned previously, more recent experimental and theoretical works show that the observed energy dispersion of the dephasing times can be explained without the need of a mobility edge [4, 24].

The three SL’s that we study in this work have been grown in the same identical condition one after the other, and thus their in-plane disorder can be assumed to be same. The aim of our work is to show that the RRS and PLE spectra are markedly different in the thee SL’s and their behaviour can be justified on the base of the different exciton localization along \( z \) due to the specific SL potential profile. To this respect, the complete theoretical analysis of the different spectra that we observe in the three SL’s is a difficult task since requires accurate models [4] and goes beyond the aim of the present work.

The main PLE peak at zero magnetic field has a different shape and width in the three SL’s, due to their different electronic structure. In particular it is wider and not symmetric in the RSL and DSL, due to their intentional disorder (see Fig. 1). This make difficult to determine the exact position of the PLE peak in the RSL and DSL and therefore to compare the shift between PLE and RRS in the three samples. However the position of the PLE edge, taken as the energy at which the PLE low energy rapid rise reaches the half value of its maximum, can be read with great precision. Besides the energy region between the RRS and the PLE edge is just the region where the dispersion of the dephasing times takes place [6, 23]. Therefore we can read the difference between the PLE edge and the RRS peak and compare it for the three SL’s. This energy difference is nearly 1 meV in the OSL whereas is close to zero for the RSL and has an intermediate value in the DSL.

This finding, i.e. the observation that the shift between RRS and PLE absorption edge of the RSL is the smaller one, is compatible with the previous interpretation and suggest that in the OSL the presence of a miniband in the \( z \) direction allows the presence of exciton with a great spectral dispersion of the dephasing rates. Within the seminal interpretation of Hegarty et al. [6, 7, 9] this corresponds to the presence of a mobility edge even if, as mentioned previously, the latest accurate theoretical models shows that the experimental RRS data and the relevant dispersion of the dephasing time can be explained on the base of accurate models without using the mobility edge concept.

In the case of the RSL the strong fluctuation of the potential profile in the growth direction due to the intentional uncorrelated disorder, induces the exciton localization along \( z \). This strong localization of the excitons in the RSL leads to a smaller shift between RRS and PLE edge then in the OSL. In the case of the DSL the presence, due to the correlation of the disorder, of a narrow band of extended states [3, 20, 21] increases the exciton mobility along \( z \) (with respect to that of the RSL) and increases the energy dispersion of the dephasing time. This gives a shift greater than the one of the RSL, but in any case smaller that than in the case of the OSL which is the SL with the greater mobility along \( z \). The DSL presents the unique characteristic of having simultaneously in the same direction \( z \) a potential profile strongly disordered together with a band of mobile states, given by the correlations. The greater value of the shift for the DSL, with respect to the shift of the RSL is a clear indication that in the DSL the presence of this extended states in the growth direction reduces the exciton localization along \( z \) influencing the spectral dispersion of the dephasing times.

In Fig. 2 we reports the same measurements of Fig. 1 but in the presence of a 14 T magnetic field applied in the growth direction and in Faraday configuration. We observe that applying a magnetic field the PLE, PL and the RRS spectra moves towards higher energies due to the diamagnetic shift of the excitons. In the meantime the HH peak widens and splits several sub-structures. Also in the presence of the magnetic field the DSL has a value of the shift that remains comprised between the values of the RSL and the OSL, being the shift of the RSL the smaller one. This indicate that the reduction of the localization given by the correlation of the disorder is a robust phenomenon. We observed the the magnetic field broadens and splits the RRS spectra in several sub-structure due to the excited states of the exciton therefore not allowing a sufficiently accurate determination of the RRS position and, as a consequence, of the shift.
PHOTOLUMINESCENCE SHIFT AND LINE WIDTH

In Fig. 3 we report the diamagnetic shift of the energy of the PL as a function of the applied magnetic field applied in the growth direction of the SL’s. The dashed line is a quadratic fit for OSL. We can observe that the energy of the main PL peak of the three samples shift to higher energy in proportion to the square of the magnetic field, as expected for excitonic transitions. The shift is nearly the same for the three SL’s and by fitting with a quadratic expression the data for the OSL (with well and barrier width of 2.6 nm) we find the diamagnetic shift to be 22 μeV/T². This value is very close to the earlier published value of 21 μeV/T² measured in QW with well width of 2.1 nm. Our finding that the diamagnetic shift of a SL is close to that of a QW with similar well width was also observed in the past by Birkedal et al. [27]. In their work they observed that a SL with well and barrier width of 8 nm and 5 nm had a shift of 26.4 μeV/T² close to the value of 29 μeV/T² measured on QW with 7.5 nm well width. Moreover our smaller value of the diamagnetic shift with respect to the one measure by Birkedal et al. [27] is coherent with the observations that the diamagnetic shift increases with the increase of the well width.

We also report in the inset of the Fig. 3 the full width at half maximum (FWHM) of the main PL peak of the three SL’s as a function of the magnetic field. All the FWHM refers to the PL measured with an excitation laser energy 0.025 eV above the PL peak position. The linewidth of the PL of the OSL increases very slightly with the magnetic field, whereas the behaviour is not monotonous in the other two SL’s. The increase of the FWHM observed in the OSL has a very similar with respect to the one measure by Birkedal et al. [27]. Moreover our smaller value of the diamagnetic shift increases with the increase of the well width.

In summary, we studied for the first time the RRS in semiconductor SL’s. This experimental technique allows to investigate in details the electron localization and transport properties of the two-dimensional electron gases in the wells. We measured ordered SL’s and intentionally disordered SL’s also with correlated disorder and we tuned the degree of localization of the excitons in the wells applying an external magnetic field. We evidence that the electronic properties along the growth direction strongly affect the mobility of the electron gases of the wells. The presence of extended states in the direction of the superlattice potential profile reduces the exciton localization in the wells and the coherence relaxation even though this potential is completely disordered.

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FIG. 1: Scattered light spectra (small points) and PLE spectra (dashed line) of the Ordered SL (a), Random SL (b) and Random dimer-SL (c) at 4 K and zero external magnetic field for $\sigma^+$ excitation and detection polarization. In the insert of panel (a) we show only some of these spectra (for which the excitation energy differs of 0.8 meV from one spectrum to the other).

FIG. 2: Scattered light spectra (small points) and PLE spectra (dashed line) of the Ordered SL (a), Random SL (b) and Random dimer-SL (c) in an external magnetic field of 14 T for $\sigma^+$ excitation and detection polarization.

FIG. 3: Energy shift of the energy of the PL for the three SL’s as a function of the magnetic field. The dashed line is a quadratic fit of the shift for the Ordered SL. The insert of panel (c) shows the full width half maximum (FWHM) of the PL peak as a function of the magnetic field.
