Recombination dynamics of spatially separated electron-hole plasmas in GaAs/AlAs mixed type-I/type-II quantum well structures

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We have studied the recombination dynamics of a series of mixed type-I/type-II GaAs/AlAs quantum well structures. While monitoring the recombination involving the wide quantum wells, we observed long-lived transients whose time scales increase exponentially with the barrier thickness of the quantum well structures. We identify heavy hole tunneling as the dominant process which determines the time scale of the long-lived photoluminescence transients. By studying the barrier thickness dependence and the excitation power dependence of the long-lived transients we have also shown that the electric field associated with the spatially separated electron/hole plasma leads to significant modifications of the relative hole subband energies and hence the hole tunneling rate.

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I. INTRODUCTION

By an appropriate design of the thickness and chemical composition of the layers of quantum well structures, it is possible to create a range of novel two-dimensional structures that exhibit specific optoelectronic properties. This is particularly the case for the GaAs/AlAs materials system where, by the choice of appropriate layer thickness, it is possible to create a so-called type-II band alignment. Such a system is created[1] when the thickness of the GaAs layer is less than 35 Å, so that the lowest confined electron state in the GaAs lies above the X minima in the AlAs. Thus the lowest energy confined electron state of the whole structure lies in the AlAs layer and the lowest energy confined hole state lies in the GaAs layer. A refinement of such a band alignment is achieved in a so-called mixed type-I/type-II structure[2] where a GaAs/AlAs multiple quantum well structure is fabricated in which alternate layers of GaAs have thicknesses greater than and less than 35 Å (see Fig. 1). The main consequence of this type of structure is that electron/hole pairs that are optically excited in the thin GaAs layer become spatially separated, as the electrons can scatter on a ps time scale[3] via the AlAs X states to the thicker GaAs layer (see Fig. 1). The spatial separation of the electrons and holes and hence long lifetime[4] means that the density of carriers can be very large ($\sim 10^{12}$ cm^{-2}) for optical excitation densities of only a few mW cm^{-2}. Initially this type of structure was used to generate large carrier densities at ultralow optical power excitation for the study[2,4] of nonlinear optical properties of the wide quantum wells. Such structures have also been used to study a wide range of physical phenomena including the behavior of charged excitons,[5–8] interlayer exciton,[9] electron/exciton scattering,[10] indirect electron-hole gas transitions,[11] and the modulation of THz radiation by an optically excited electron gas.[12]

In the original reports[2,4] on the mixed type-I/type-II structure it was inferred from the dependence of the density of the electron gas on the barrier thickness that the lifetime of the spatially separated carriers was determined by the heavy hole tunneling time. The heavy hole tunneling times were estimated from the recovery time of the exciton bleaching following pulsed excitation. The times quoted in this work could only be regarded as estimates because the nature of the response of the exciton bleaching as a function of carrier density was not known. In this paper we report on the observation, following pulsed excitation, of long-lived recombination from the wide quantum well, which we ascribe to the sequential process of holes tunneling from the narrow quantum well followed by recombination with electrons in the wide quantum well. These observations are used to investigate the dynamics of hole tunneling as a function of carrier density in samples with a range of barrier widths.

II. EXPERIMENTAL DETAILS

The samples were grown by molecular beam epitaxy in a Varian Gen II machine at a substrate temperature of 630 °C

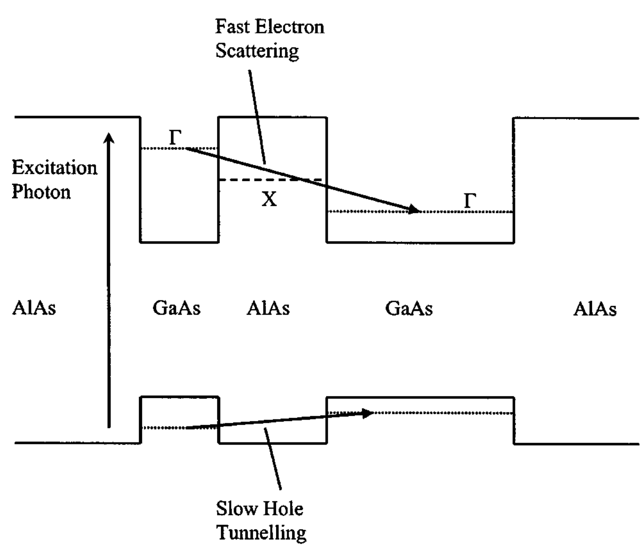

FIG. 1. Schematic diagram of the conduction and valence band edges showing the lowest confined X and Γ electron states and hole states. The dominant excitation mechanism and the limiting electron and hole relaxation mechanisms are indicated by arrows.

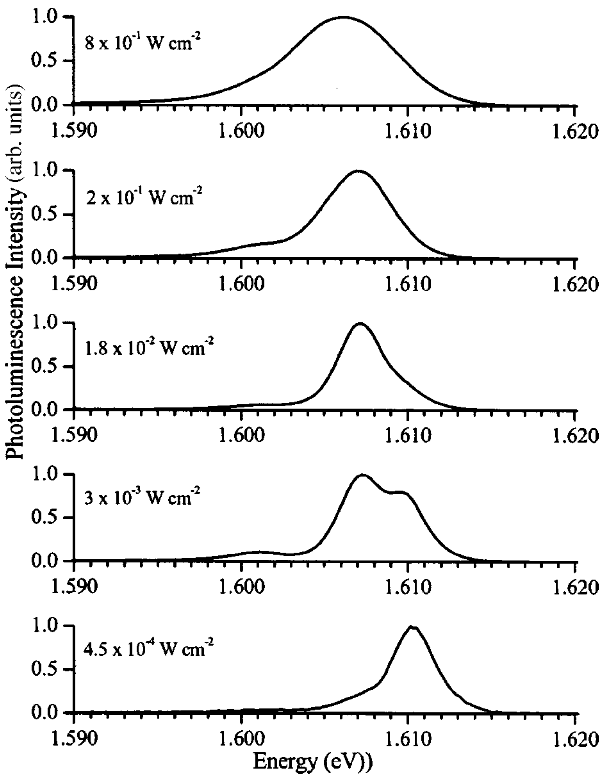

FIG. 2. Low temperature ($T=8$ K) photoluminescence spectra from the sample with 80-Å barriers as a function of the average excitation power density.

using As_2. The samples were grown on n^+ substrates and consisted of nominally (a) 6800 Å of GaAs, (b) a 6000-Å-thick layer of $Al_{0.6}Ga_{0.4}As$, (c) a multiple quantum well consisting of 20 periods of 25- and 68-Å GaAs double quantum wells separated by AlAs barriers of width 72, 80, 90, and 109 Å in the different samples, (d) a 6000-Å-thick layer of $Al_{0.6}Ga_{0.4}As$, and (e) a 1000-Å-thick GaAs capping layer. The thicknesses of the GaAs and AlAs layers were measured[4] by x-ray diffraction.

For the optical experiments the samples were mounted on the cold finger of a liquid He continuous flow cryostat. For the cw photoluminescence spectroscopy the samples were excited by chopped light from a He/Ne laser and the emission analyzed and detected by a 1-m grating spectrometer followed by a GaAs photomultiplier and lock-in amplifier. The photoluminescence time decay measurements were performed by modulating the He/Ne laser with an acousto-optic modulator and processing the photoluminescence signal with a digital oscilloscope.

III. RESULTS AND DISCUSSION

In Fig. 2 we show a series of photoluminescence spectra recorded at a temperature of 8 K as a function of excitation power density from the sample with 80-Å barriers. At the lowest excitation power density the spectrum is dominated by a feature at 1.6104 eV, which is ascribed to exciton emission involving the lowest confined electron and hole states of the wide quantum well. As the excitation power density is increased, a low energy shoulder emerges at 1.6073 eV, which is identified as recombination involving negatively charged excitons,[5] indicating a significant free electron density. The feature at 1.6011 eV is identified as exciton recombination in regions where the quantum well is thicker than the average, which may be within a quantum well or in quantum wells with different thickness; this assignment is based on absorption spectroscopy (not shown). As the excitation power density is increased, the individual peaks can no longer be resolved and the spectrum broadens reflecting the large electron density that exists in the wide quantum well. The reasons why the values of excitation power density are quoted and not the equivalent excited carrier densities will be discussed in detail later. These spectra are typical of the results from all four samples, the only difference being the form of the spectra is different for a specific excitation power density, which reflects the dependence of the electron density in the wide quantum well on the barrier thickness.

The basic idea behind this work is that if the decay of the photoluminescence from the wide quantum well is monitored following pulsed excitation then not only will we observe the fast (∼ns) recombination of carriers excited directly in the wide quantum well but also we should see a much slower transient. This slow decay should arise from the sequential process of hole tunnelling and carrier cooling and subsequent recombination with electrons that have scattered rapidly from the narrow quantum well. The time constant associated with this sequential process will be governed by the hole tunnelling rate which is expected to be much less than the rates associated with scattering and recombination. In Fig. 3 we show decay curves taken at different detection energies across the spectrum of the sample with 80-Å barriers (photoluminescence spectra shown in Fig. 2). The sample was excited by pulses of 400 μsecs duration. In Fig. 3 slow decays can clearly be seen after the fast decay of the directly excited electron hole pairs; the dashed line in the separate decay curves indicates the turn-off time of the excitation light. The measured decay of the initial fast transient is limited by the system response. These results illustrate one of the main complications in performing photoluminescence time decay measurements in this type of sample. If the decay of the photoluminescence is measured at a particular detection energy, then the form of the decay will not only involve the change in carrier density as a function of time but also will inevitably involve the change in the spectrum, because as seen in Fig. 2 the spectrum is a strong function of the carrier density. Thus, this method cannot be easily used to monitor the change in electron density in the wide quantum well as a function of time.

To overcome this problem, the decay of the photoluminescence was measured while sampling all the light emitted by the samples. Control experiments were performed that showed there was no significant contribution to the signal from recombination involving electrons and holes in the narrow quantum well, electrons in the X states in the AlAs barriers and holes in the narrow quantum well and electrons and holes in the bulk GaAs. Thus, we are able to monitor the change in carrier density in the wide quantum well as a function of time. In Fig. 4 we show the results of the all-light

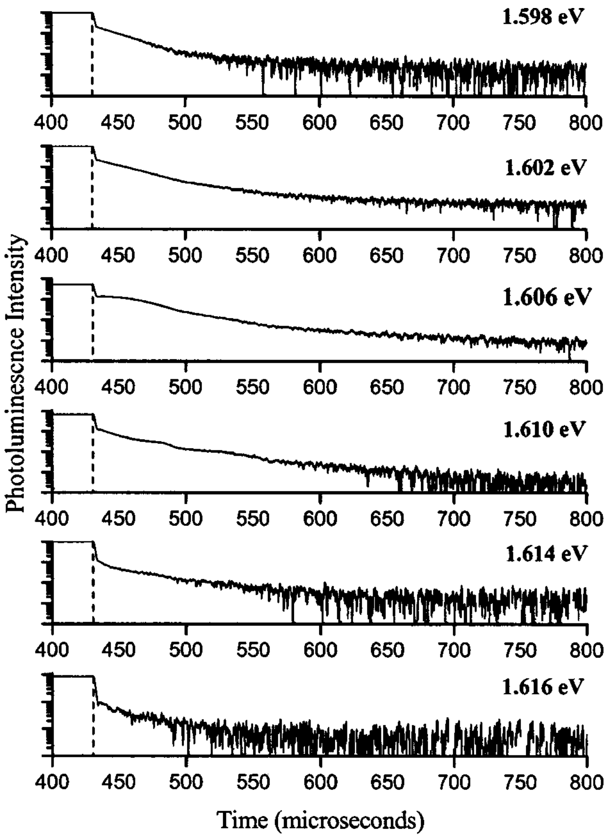

FIG. 3. Photoluminescence transients recorded at $T=8$ K from the sample with the 80-Å barriers at various detection energies across the spectrum for an average excitation power density of 8×10^{-1} W cm^{-2}.

decay time measurements for all four samples where the intensity of the excitation pulse was varied as indicated in the figure caption. All the decay curves were normalized such that the maximum intensity at the end of the excitation pulse was set equal to 1, and then offset for clarity.

Clearly, the form of the decay curves varies with excitation power density and from sample to sample at the same excitation power density. To clarify the overall behavior we start by noting that for all four samples at the lowest excitation power density the decays have the same general nonexponential form. As stated previously we attribute these long-lived transients to the holes tunneling from the narrow quantum well to the wide quantum well. As these decay curves are nonexponential we characterize them by the time (τ_e) for the photoluminescence intensity to fall from its maximum value at the beginning of the long decay to a value of $1/e$ of the maximum value. The values for τ_e are listed in Table I and, as can be seen, they increase rapidly as a function of the barrier thickness. It should be stressed that the values of τ_e have little physical significance other than lending support to the assignment of the process controlling the slow transient as being the hole-tunneling rate. Nevertheless, this argument is supported by the data presented in Fig. 5, where the values of τ_e are shown on a log/linear plot as a function of the barrier thickness and a reasonable exponential dependence is observed. In the simplest picture, one would expect that the decays at the lowest excitation power density would be governed by a single time constant, i.e., single exponentials. We suggest that the observed nonexponential character of the slow decay curves at low excitation densities is due to interface roughness. As the hetero-

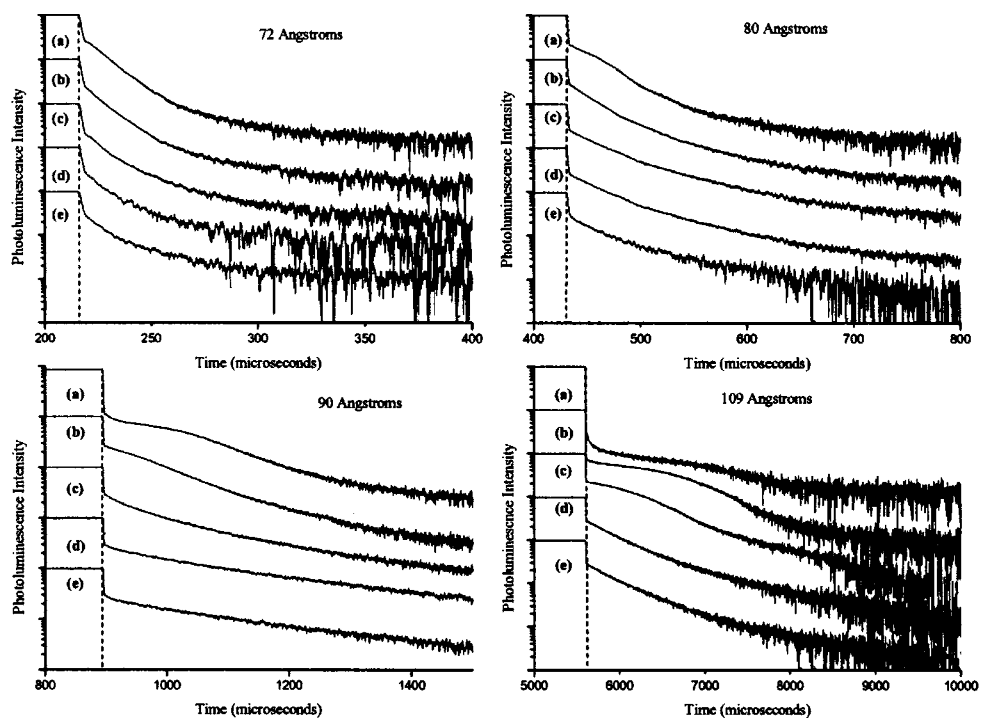

FIG. 4. Photoluminescence transients of all the light from all four samples with the indicated barrier thickness at different excitation power densities. For the curves labeled (a), (b), (c), (d) and (e) the excitation power densities were 1.4, 1.3×10^{-1}, 1.3×10^{-2}, 1.2×10^{-3}, and 6×10^{-4} W cm^{-2}, respectively.

TABLE I. For all four samples are listed values of τ_e, the area under the decay curve, the estimated value of the carrier density n and the relative shift in energy of the hole subbands ΔE, all in the low excitation region.

Power density (W cm^{-2})	τ_e (microsecs)	Integrated area (a.u.)	n (cm^{-2})	ΔE (meV)
Barrier thickness = 72 Å				
1.4				
1.3×10^{-1}	11	3	10^{10}	1.2
1.3×10^{-2}	11	3	10^{9}	0.1
1.2×10^{-3}	13	3	10^{8}	0.01
6×10^{-4}	13	3	5×10^{7}	0.005
Barrier thickness = 80 Å				
1.4				
1.3×10^{-1}				
1.3×10^{-2}	38	11	7×10^{9}	0.8
1.2×10^{-3}	36	12	1×10^{8}	0.08
6×10^{-4}	34	12	3×10^{8}	0.004
Barrier thickness = 90 Å				
1.4				
1.3×10^{-1}				
1.3×10^{-2}				
1.2×10^{-3}	172	60	5×10^{9}	0.8
6×10^{-4}	176	62	3×10^{8}	0.4
Barrier thickness = 109 Å				
1.4				
1.3×10^{-1}				
1.3×10^{-2}				
1.2×10^{-3}	422	133	3×10^{9}	5
6×10^{-4}	438	123	1.5×10^{9}	2.5

interfaces of the quantum wells are not atomically smooth, there are local fluctuations in the thickness of the AlAs barriers. Thus the holes will tunnel through barriers of different thickness, leading to a wide range of tunneling rates within the excitation area.

As the excitation power density is increased the form of the decay curves diverges from that exhibited in the subset of curves labeled (e) in Fig. 4. As an example, if we examine the data for the sample with 90-Å barriers we see that [curve (b)] the decay initially exhibits a portion with a relatively small gradient before decaying at a similar rate to that in curve (e). Furthermore, at the highest excitation power density [curve (a)], the decay exhibits an initial fast transient before the flatter portion of the decay curve is reached. All the samples show a similar behavior with increasing excitation power density, although it should be noted that (a) the onset of the flat region of the decay occurs at lower excitation power densities as the barrier thickness increases, and (b) the fast transients observed at the high excitation power densities become significant only in the samples with barrier thicknesses of 90 and 109 Å. The dependence of these two characteristics on both excitation power density and barrier thickness, suggests that the controlling factor is the equilibrium carrier density achieved during the pump pulse. As pointed out previously[4,11] the spatially separated electron hole plasmas can lead to a significant space charge field. This electric field can lead to significant changes in the alignment of the hole subbands in the narrow and wide quantum wells. Results for the calculated dispersion[13] of the valence subbands for the two quantum wells are shown in Fig. 6 in the absence of any space charge field. The most important sub-

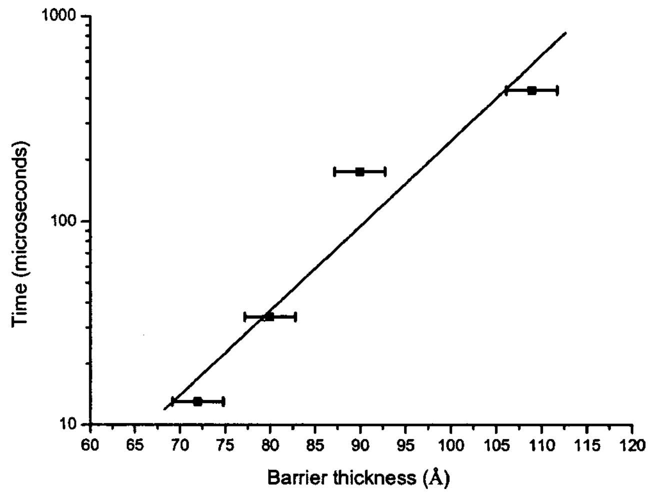

FIG. 5. A plot of τ_e vs barrier thickness for all four samples, an uncertainty in the barrier thickness of ± one monolayer is assumed.

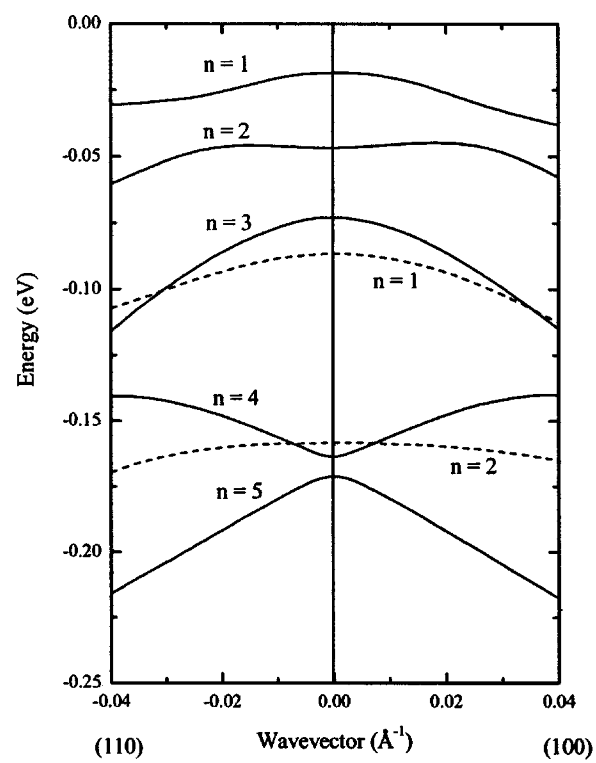

FIG. 6. Valence subbands for the 25- and 68-Å GaAs/AlAs quantum wells indicated by the dashed and solid lines, respectively. The labeling of the states is made without any consideration to their heavy or light hole character.

bands for the purposes of this discussion are the lowest lying $n=1$ subband in the 25-Å quantum well and the third and fourth highest bands in the 68-Å quantum well. The dominant effect of a space charge field will be to raise the lowest energy 25-Å hole subband relative to the subbands in the wide quantum well. Thus, as any space charge field is increased the intersection of the $n=1$ subband in the 25-Å quantum well with the $n=3$ hole subband of the wide quantum well will shift to increasingly large values of k, for which the thermal factor is smaller, leading to a reduction in the tunnelling current. Eventually the $n=1$ hole subband of the 25-Å quantum well will come into resonance with the $n=4$ hole subband of the 68-Å quantum well. Qualitatively this overall behavior agrees with the experimental observations, in which for the samples with the 90 and 109-Å barriers at the highest excitation power densities we see an initial fast transient followed by a relatively flat portion of the decay curve. These effects are seen in these two samples in particular because the overall tunnelling rates are low, leading to the high carrier densities and hence the large space charge fields necessary.

It is difficult to put these arguments on a quantitative basis, as we cannot use any single decay rate to calculate the equilibrium carrier densities. Nevertheless a rough estimate of the carrier densities (n) and hence the relative shift (ΔE) in the hole subband energies can be obtained for the low excitation power densities by using the observed values of τ_e. The values for the estimated carrier densities at the low excitation power densities are shown in Table I. For the decay curves from which the values of τ_e were extracted, the relative shift of the hole subbands is no more than a few meV.

If these time constants were appropriate for the excitation power densities used for the decay curves that show the initial fast transient and the flat decay the carrier densities would be sufficient to produce changes in the alignment of the hole subbands of $\approx$100 meV. This would lead to the $n=1$ hole subband of the 25-Å quantum well being in or close to resonance with the $n=4$ hole subband of the wide quantum well leading to an enhancement of the hole tunnelling rate. As the carrier density collapses the $n=1$ hole subband of the 25-Å quantum well no longer intersects with the $n=4$ wide well subband, but instead intersects with the third wide well subband at high k values leading to a reduction in the hole tunnelling rate and the shallow portion region of the decay.

The preceding discussion assumes that the decays we measure are purely radiative, i.e., that there are no nonradiative recombination paths. As these systems exhibit such long-lived carriers they could be particularly susceptible to nonradiative recombination. To investigate this possibility we consider the integrated area under the decay curves for the low excitation power densities. Based on the argument that the values of τ_e are a measure of the decay of the spatially separated electron/hole pairs, the magnitude of the integrated areas under the decay curves should scale with the values of τ_e for the different samples if the decays are purely radiative. The values for the integrated areas are shown in Table I, and indeed, at the low excitation densities the integrated areas do scale with τ_e. Thus, we believe that the effects of nonradiative recombination are negligible even over these long time scales.

IV. SUMMARY

In summary, we have observed complex long-lived photoluminescence transients involving recombination from electrons and holes in the wide quantum well of a variety of mixed type-I/type-II quantum well structures. By studying the barrier dependence of the form and time scale of the long-lived transients we have shown that the dominating factor is rate at which holes tunnel from the narrow quantum wells to the wide quantum wells. In particular the form of the transient is strongly influenced by the relative alignment of the hole subbands in the quantum wells which itself is strongly influenced by the time dependent space charge field caused by the spatially separated electron hole plasma.

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