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Femtosecond pump-probe spectroscopy investigation of carrier dynamics in GaAlAs photocathods

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ABSTRACT

Based on the study of GaAs photocathode, the carrier dynamics of reflection-mode GaAlAs photocathode has been investigated. In this paper, the reflection-mode GaAlAs/GaAs photocathode is grown by metal organic chemical vapor deposition (MOCVD) epitaxial technology. The femtosecond pump-probe spectroscopy is performed to measure carrier dynamics in GaAlAs and GaAs photocathods. After laser excitation the surfaces of GaAlAs/GaAs photocathode, the reflectivity of photocathode surface is changed. At the same time, the dynamic processes of the non-equilibrium carrier distribution in semiconductor material is obtained by the transient reflectance spectrum. Compared with GaAs photocathode, the Al element in GaAlAs photocathode is unfavorable to photoemission under some conditions, while it solves the problem that the spectrum response range of binary compounds is not adjustable. The change trend of initial relaxation transient of GaAs photocathode in the 22 ps range is the same as GaAlAs, while the change of GaAs and GaAlAs relaxation state is significantly different with the change of delay time after 22 ps. The results show that the GaAlAs increases band gap due to a significantly slower than GaAs photocathode in initial process. Meanwhile, this paper provides a reference for the preparation of the GaAlAs photocathode that only respond to blue and green light range.

1. Introduction

As the core component of low-light-level image intensifier, III-V GaAlAs and GaAs have already found widespread application in optoelectronic devices such as image intensifiers, photomultipliers, polarized electron source and solar cell [1–4]. In GaAs photocathodes, the GaAlAs is used as the buffer layer which have a good lattice match with the high-grade n-GaAs substrate. What's more, the GaAlAs can absorb photons and participate in the final photoemission [5]. The GaAlAs photocathodes may be activated by adsorbing Cs/O on the clean surface and form the negative electron affinity (NEA) photocathodes [6,7]. As we know, the response wavelengths are different for the different photocathodes and the application of GaAs photocathode is limited thanks to the fixed band gap and the wide spectral response range [7,8]. However, when putting into Al element or changing the Al fraction in GaAs, the spectral response range changes accordingly. The three-step process is used to investigate the NEA photocathodes photoemission which includes optical absorption, electron transport and electron escape [9]. According to the three-step process, the carrier energy distribution and the quantum efficiency are two important sides on the photoemission of NEA photocathode.

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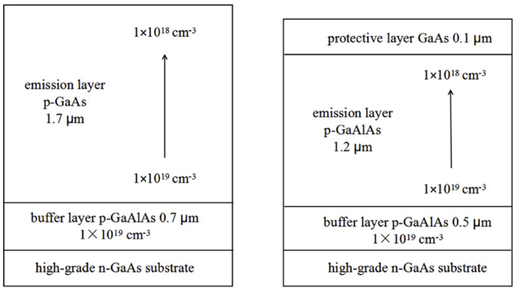

(a) GaAs (b) GaAlAs

Fig. 1. Structure of exponential-doping reflection-mode (a) GaAs and (b) GaAlAs photocathodes sample.

The dynamics of excited carriers are related to both electronic and optoelectronic for GaAlAs or GaAs photocathode. Using the laser excites the surfaces of GaAlAs or GaAs photocathode respectively, the reflectivity of photocathode surface is changed. In addition, the dynamic processes of the non-equilibrium carriers distribution in semiconductor materials are analyzed by researching the transient reflectance spectrum. Shank et al. Use continuum pump-probe technique to examine state filling and band renormalization [10,11]. By comparing the macroscopic quantum efficiency and absorption coefficient curves of the exponentially doped reflection-mode GaAlAs and GaAs photocathodes, it is found that the addition of Al component is not conducive to photoemission to some extent. However, it solves the problem that the response band of GaAs photocathode is wide and unadjustable. In this letter, combined with the microcosmic method of femtosecond pump-probe spectroscopy, the macroscopic phenomena has been explained and make it possible to study the transient properties of semiconductor materials from microscopis [12–15]. This paper mainly introduces the method to study GaAlAs/GaAs carrier relaxation process which performs a two-pulse correlation way. It provides new information for improving the performance of materials and devices and also gives a reference for preparation the GaAlAs photocathode on responding blue and green light.

2. Experimental

Experimental measurements are performed on high-grade n-GaAs substrates of GaAs and GaAlAs with epitaxial layer grown by MOCVD epitaxial technology. The emissive layer of both photocathodes are exponentially doped and these measurements are performed for perpendicular polarization of pump and probe beams. Fig. 1 shows the structure of exponential-doping reflection-mode GaAs and GaAlAs photocathodes. Moreover, the Al content of the emission layer in the GaAlAs photocathode sample is 0.5. A GaAs protective layer is grown on the (b) GaAlAs photocathode emission layer in case the GaAlAs material is oxidized in the air.

The photocathode forms a bipolar layer which has NEA after activation and doping. The energy band structure of exponential-doping GaAlAs photocathode is shown in Fig. 2. And the exponential doping structure results in built-in electric field in the photocathode body, which causes the band to form a structure inclined from the body to the surface and it improves the photoemission performance of the reflection-mode photocathode [16]. When a femtosecond pulse interacts with the photocathodes, electron and hole pairs are first generated by interband absorption. At the same time, the excited carriers occupy some states which increase reflectivity and generate saturation absorption effect. And the energy distribution of the excited carriers is determined by the pulse energy of incident light. With the recombination of electrons and holes, the saturation effect is weakened and the reflectivity decreases. And the measurements are all performed at room temperature to excite and probe the reflectivity change. The microscopic dynamic process of GaAlAs photocathode may be gained by detecting the transient reflectance spectrum on the surface of the photocathode. Then the carrier relaxation can be arised by several different mechanisms [17]. Moreover, carrier-carrier scattering

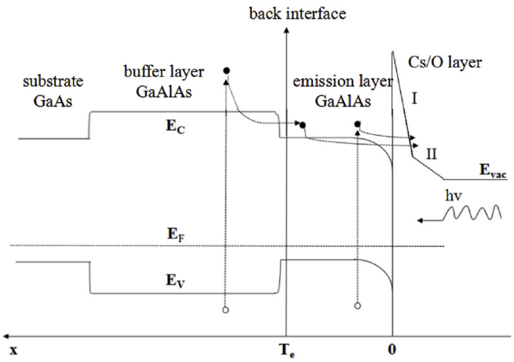

Fig. 2. The energy band structure of exponential-doping GaAlAs photocathode. E_V is the maximum value in valence band, E_F is the Fermi level, E_C is the minimum value in conduction band, E_{vac} is the vacuum level and hv is the incident light energy.

and carrier-phonon scattering build up the thermal equilibrium together. Carriers reduce their energy by spontaneously generating phonon which transfer energy to the lattice, while the number of carriers does not decrease.

3. Results and discussion

The quantum efficiency is an important characteristic parameter which able to characterize the photoemission properties of reflection-mode GaAlAs/GaAs photocathode. Usually, the experimental curve is fitted by quantum efficiency formula, and the related performance parameters can be obtained according to the quantum efficiency curve.

And the quantum efficiency formula of reflection-mode GaAlAs and GaAs photocathodes are as follow [18]:

$$Y_{RE} = \frac{P(1-R)\alpha_{h\nu}L_D}{\alpha_{h\nu}^2 L_D - \alpha_{h\nu}L_E - 1} \times \left[\frac{N(S-\alpha_{h\nu}D_n)e^{\frac{L_E T_e}{2L_D^2} - \alpha_{h\nu}T_e}}{M} - \frac{Q}{M} + \alpha_{h\nu}L_D \right] \tag{1}$$

where

$$N = \sqrt{L_E^2 + 4L_D^2}$$

$$S = \mu\ |E| + S_\nu$$

$$L_E = \mu\ |E|\ \tau = \frac{q\ |E|}{k_0 T} L_D^2$$

$$M = \left({}^{ND_n}\!/_{L_D} \right) \cosh\left({}^{NT_e}\!/_{2L_D^2} \right) + \left(2SL_D - {}^{D_n L_E}\!/_{L_D} \right) \sinh\left({}^{NT_e}\!/_{2L_D^2} \right)$$

$$Q = SN\cosh\left({}^{NT_e}\!/_{2L_D^2} \right) + (SL_E + 2D_n)\sinh\left({}^{NT_e}\!/_{2L_D^2} \right)$$

where Y_{RE} is the quantum efficiency, P is the probability of surface electron escape, R is the reflectivity of photocathode surface, $\alpha_{h\nu}$ is the absorption coefficient, L_D is the electron diffusion length in the GaAlAs layer, L_E is the electron drift length under an internal electric field E, τ is the lifetime of minority carrier, T is the absolute temperature, q is the electron charge, N is the doping concentration, D_n is the electron diffusion coefficient of, T_e is the thickness of GaAlAs/GaAs photocathode emission layer.

What's more, investigating the effect of these parameters on quantum efficiency, it illustrate the difference of photoemission performance between GaAlAs and GaAs photocathodes. Of course, the spectral response also can be research photoemission for the NEA photocathode and it turns out that the surface electron escape of GaAlAs photocathode is lower than GaAs due to the Al element [19]. The exponential-doped reflective GaAlAs and GaAs photocathodes quantum efficiency curves are shown in Fig. 3. And the Fig. 3 shows that the peaks of quantum efficiency of the two photocathodes appears in the high energy region, while with the increase of the incident energy, the growth rates of GaAlAs and GaAs photocathodes quantum efficiency all decrease. The reason why is that the optoelectrons escape from surface to vacuum having short distance and consuming less energy. When the incident light directly acts on the emission layer for the reflection-mode photocathode, the high-energy photons mainly absorb energy at the near surface and then produce optoelectrons. And it may be observed that the spectral response range is narrowed as the Al component adding into GaAs photocathode. Of course, the spectral response range becomes adjustable with the different Al.

The absorption coefficient of reflection-mode GaAlAs and GaAs photocathodes emission layers follows the change of the incident light energy, as shown in Fig. 4. From the Fig. 4, the absorption ability of GaAs photocathode to the incident light energy is higher than that of GaAlAs for the same energy in centain range. Moreover, with the incident energy increase, the absorption coefficients are becoming more and more different.

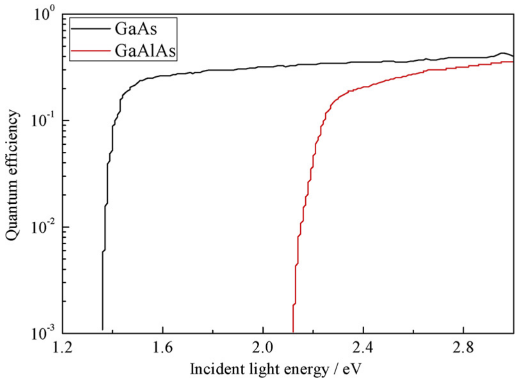

Fig. 3. The curves are exponential-doped reflective GaAlAs and GaAs photocathodes quantum efficiency after fitting.

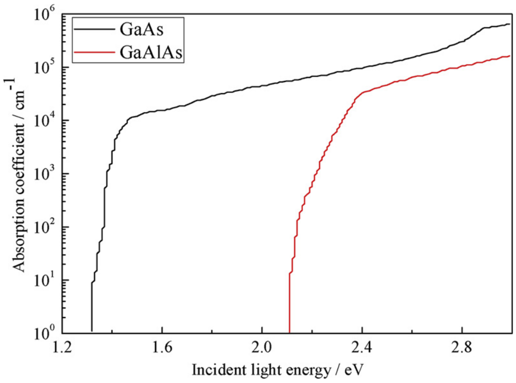

Fig. 4. The absorption coefficient curves of exponential-doped reflection-mode GaAlAs and GaAs photocathodes emission layer.

In general, it is found that GaAs photocathode is more favorable than GaAlAs in the sides of absorbing optical energy and quantum efficiency, while the applicable wavelength range is limited and wider of GaAs. Fig. 5 shows a schematic of the transient reflectance change of reflection-mode GaAlAs and GaAs photocathodes. The ΔT represents the normalization of the probe reflectivity of the photocathode sample surface with different delay time. When the GaAlAs/GaAs photocathode sample is excited by a femtosecond laser pulse, the excited electrons in the conduction band and holes in the valence band occupy some energy states. And this reduce the sample's absorption of the probe light, resulting in maximum reflectivity. However, with the attenuation of the excited carriers population, the effect degrades and the reflectivity of probe decreases. As be seen Fig. 5, in the initial delay time, the GaAs reflectivity is smaller than GaAlAs. This phenomenon means that the GaAs has a higher capacity than GaAlAs for incident light in the start phase, which corresponds to the absorption coefficient and quantum efficiency curves. It is interesting to note that the GaAlAs is rapidly relaxed within 8 ps–24 ps with the delay time increases, and then the relaxation rate becomes slow. Before 22 ps, GaAlAs and GaAs photocathodes show similar changes in the reflectivity of probe light during the corresponding delay time, while a larger phase difference after 22 ps. The initial rapid relaxation process observes in GaAlAs photocathode as well as its pronounced carrier density and also attributes the success to the split-off transition. The phenomenon can be interpreted as that the excited carriers are generated in the band edge as lower energy carriers and the influence of the nonequilibrium carriers are also considered. The quantum efficiency and absorption coefficient of exponential-doped reflection-mode GaAlAs and GaAs photocathodes are analyzed by femtosecond laser microanalysis. On the one hand, the absorption efficiency of GaAs is better than GaAlAs, while is limited for responding to the spectrum. On the other hand, with the increase of the delay time, the relaxation process of GaAlAs is slower than that of GaAs which is the better lattice match and post interface recombination.

4. Summary

In the paper, a comparative study of exponential-doped reflection-mode GaAlAs and GaAs in macro and microcosmic method has been put forward. Based on the research of GaAs photocathode, femtosecond pump-probe spectroscopy investigates the exponential-doped reflection-mode GaAlAs photocathode. Using laser excites GaAlAs/GaAs photocathode, then the surface reflectivity makes a difference. The macroscopic quantum efficiency and absorption coefficient have been performed for the GaAlAs and GaAs photocathodes and it is can be found that Al component is not conducive to photoemission in some cases, while it makes it possible to adjust the range of the wavelength response. From the microcosmic investigation of GaAlAs and GaAs photocathodes, the initial relaxation transient are same within 22 ps, while the relaxation process is slower of GaAlAs than GaAs after 22 ps which is the better

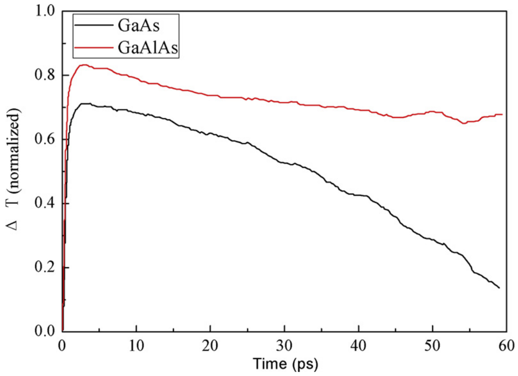

Fig. 5. The transient reflectivity change of exponential-doped reflection-mode GaAlAs and GaAs photocathodes.

lattice match and post interface recombination. Moreover, the initial rapid relaxation have been observed in GaAlAs photocathode as well as its pronounced carrier density and also attributed it to the split-off transition. And the reason why is that the excited carriers are generated in the band edge as lower energy carriers and considering the effect of the nonequilibrium carriers. In this paper, a new analytical and research method is proposed to improve the performance of GaAlAs photocathode, which make it possible to prepare GaAlAs photocathode that only responds to bule and green light.

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