Recombination radiation of electron-hole liquid with plasmon participation in uniformly deformed germanium

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A new emission line (LP), which is a satellite of the emission line of an electron-hole liquid (EHL), was observed in the emission spectrum of a pure uniaxially compressed germanium crystal. The behavior of the new line at various strains and temperatures and in a magentic field are explained in terms of recombination of electron-hole pairs in the EHL, accompanied by plasmon emission. The equilibrium density of the EHL at various values and directions of the strains is determined from the spectral position of the LP line.

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§1. INTRODUCTION

It is known that an electron-hole liquid (EHL), comprising a two-component electron-hole plasma, is produced in the indirect semiconductors Ge and Si at low temperatures.[1] There are various methods of studying the plasma properties of the EHL, one being the study of the resonant absorption in the far IR. Investigations of this kind were performed in underformed Ge and made it possible to determine for the first time the equilibrium density of the EHL in it.[2] The plasma properties of an EHL should manifest themselves spectroscopically in a deviation of the shape of the EHL emission line (L) from the shape calculated in the framework of the single-particle description. Recall that the shape of the L line is determined by transitions into excited states, which are, in the single-particle description, holes within the Fermi sea of electrons and electrons within the Fermi sea of holes. Allowance for multiparticle effects leads to the appearance of a low energy tail of the L line, as well as to an additional emission line separated from the L line by a distance equal to the plasmon energy. These deviations are due to the fact that additional excited quasiparticles and plasmons appear in the final state.[3–6] The deviations of the EHL emission-line shape from the shape calculated in the single-particle description were observed experimentally in Ge in the form of a low-energy tail of the L line.[1] Nonetheless, no detailed spectroscopic investigations of this tail were made, partly because its intensity in undeformed Ge does not exceed 5% of the total intensity, and partly because it contains another superimposed phonon replica (the difference between the energies of the TO and LA phonons is close to the width of the L line in underformed Ge).

In Ge deformed along the $\langle 111\rangle$ axis (Ge $\langle 111\rangle$) and along an axis close but not equal to $\langle 100\rangle$[1)] (Ge $\langle 100\rangle$), where the stability and equilibrium density of the EHL decrease significantly as a result of the decrease of the state densities of the electrons and holes in the bands, the role of multiparticle effects in the formation of the emission spectrum becomes much greater than in undeformed Ge.[7] This leads to an appreciable deviation of the shape of the L line from that expected within the framework of the single-particle description, and consequently to an appreciable error in the determination of the equilibrium density of the EHL by approximating the shape of the L line.

We have investigated experimentally the role of multiparticle effects in the formation of the emission spectrum of the EHL in deformed Ge. In addition to the detailed investigation of the red tail of the L line (§3) we have observed an additional emission line (§§3,5) due to the appearance of a plasmon in the final state. From the position and shape of the additional line we determined the equilibrium density of the EHL at different values and directions of the strain (§4).

§2. EXPERIMENTAL PROCEDURE

We investigated Ge with shallow-impurity density $N \approx 3\times 10^{12}\,\mathrm{cm}^{-3}$. To decrease the state densities of the electrons and holes in the bands we used uniaxial compression of crystals measuring $3\times 3\times 10$ mm, which were first polished mechanically and then chemically by etching in CP-4a. The procedure of obtaining large uniform compression of the crystals was described earlier.[7] The samples were located in superfluid helium, so that their heating by optical excitation was a minimum. The nonequilibrium carriers were excited with a cw aluminum-yttrium-garnet laser ($\lambda = 1.064\,\mu$m) of up to 6 W power.

The spectral instrument was a double monochromator with a grating of 600 lines/mm and dispersion 8 Å/mm. The radiation was registered with a Ge(Cu) photoresistor cooled to liquid-nitrogen temperature. The magnetic field was always parallel to the strain axis. The radiation was recorded in the Voigt geometry ($\mathbf{H}\perp\mathbf{k}$).

§3. EMISSION SPECTRA OF STRONGLY COMPRESSED Ge $\langle 100\rangle$ CRYSTALS

Within the framework of the single-particle description, the L-line shape is determined by transitions from the ground (GS) to the excited state:

$$\mathrm{GS} \to \tilde{e} + \tilde{h} + h\nu + \hbar\Omega, \tag{1}$$

where $\tilde{e}(\tilde{h})$ denotes the single-particle excitation [electron (hole) in the Fermi sea of the holes (electrons)], $h\nu$ is the emitted photon, $\hbar\Omega$ is the phonon needed to satisfy the quasimomentum conservation law. In undeformed Ge, where

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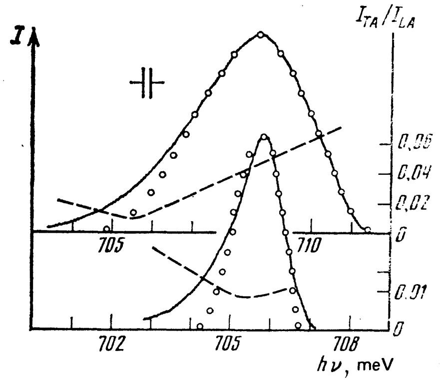

FIG. 1. Allowed component of the EHL emission line (L) in undeformed (upper part of the figure) and strongly deformed Ge ⟨100⟩ ($P \approx 470$ MPa, lower part of the figure) at $T = 1.7$ K. The L-line shape calculated in the single-particle approximation is shown by the circles. The dashed lines show the ratio of the forbidden (TA) and allowed emission-spectrum components made congruent by shifting the TA component by a distance equal to the energy difference of the LA and TA phonons.

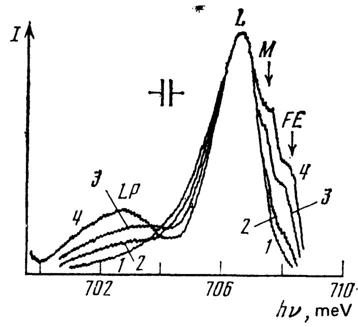

FIG. 2. Emission spectra of Ge ⟨100⟩ at $P = 420$ MPa, $T \approx 1.5$ K, and various excitation densities (W): 1–200, 2–120, 3–100, 4–70 W/cm^2.

the deviation of the L-line shape from the single-particle description is small, the low-energy tail contains about 5% of the total intensity of the L line (Fig. 1). This deviation was usually attributed to damping of the indicated single particle excitations. In that case this assumption was valid, since the damping γ needed for a good description of the red tail of the L line was small compared with the carrier Fermi energies.[8] In the case of Ge ⟨100⟩, where the relative integral intensity of the red tail of the L line increases to 20% (Fig. 1), the L-line shape cannot be satisfactorily described even at values of γ exceeding the Fermi energies of the electrons and holes. This is evidence that the assumption made above concerning the damping of single-particle excitations does not hold in the case of the low density of the EHL in Ge ⟨100⟩.

On the other hand, as noted in §1, another approach can describe the low-energy tail of the L line, by attributing it to the appearance of additional excited quasiparticles and plasmons in the final state, i.e., to process of the type[5,6]

$$\mathrm{GS} \rightarrow \tilde{e} + \tilde{h} + h\nu + \hbar\Omega + X, \qquad (2)$$

where X denotes the additional recoil particle, which can be either a plasmon ($X = \hbar\omega_p$) or an electron-hole pair produced upon excitation of an Auger electron (e) outside the Fermi sphere: $X = e + \tilde{h}$ ($X = h + \tilde{e}$ in the case of a hole).

In Ge, where the radiation is indirect and is accompanied by emission of a phonon that carries away quasimomentum in accord with the conservation law, there is a method of separating recoil processes of type (2) from single-particle processes (1). In this method one compares the spectra of the radiation with emission of LA and TA phonons, which correspond, in accord with the symmetry laws, to transitions allowed and forbidden in zeroth order in **k**. It is known that recoil processes are typified by a negative slope of the plot of the spectrum ratio in the TA and LA components (I_{TA}/I_{LA}) vs the energy of the emitted photon,[9] so that to detect process of type (2) it is necessary to study the $I_{TA}/I_{LA}(h\nu)$ dependences. Figure 1 shows plots of $I_{TA}/I_{LA}(h\nu)$ for EHL in undeformed Ge and in Ge ⟨100⟩, from which it can be directly seen that the low-energy tail of the line L is due to recoil processes of type (2), whose probability increases with decreasing EHL density.

Investigation of the shape of the L line has shown that at large strains ($P \gtrsim 300$ MPa), at the lowest temperatures ($T \lesssim 2$ K) and under definite focusing conditions the red tail of the L line is transformed into a separate well-resolved line LP. Figure 2 demonstrates the change of the radiation spectrum as a function of the focusing conditions at $P = 400$ MPa and $T = 1.8$ K (the excitation-spot size changes at a constant laser power). Spectrum 1 in this figure corresponds to maximum focusing with power density $W_1 = 200$ W/cm.^{2} With decreasing excitation density the red tail of line L is transformed first into a separate shoulder ($W_2 = 120$ W/cm^2), and then into the resolved LP line ($W_3 = 100$ W/cm^2, $W_4 = 70$ W/cm^2). With further decrease of the excitation density ($W < 70$ W/cm^2) the relative intensity of the LP line ($\alpha = I_{\mathrm{LP}}/I_L$) decreased somewhat. It should be pointed out that the dependence of the emission spectra on the excitation density, shown in Fig. 2, cannot be recommended as a general method of obtaining an intense LP line, since it was necessary to choose for the different samples different optimal values of the power density W_0 (in the different samples W_0 ranged from 60 to 120 W/cm^2 and α from 0.15 to 0.30). It seems therefore that the relative intensity of the LP line depends not only on the external parameters (temperature and strain) but also on "internal" parameter that is very difficult to control. Such a parameter can be, e.g., the size of the electron-hole drops (see §5).

To ascertain the nature of the LP line we used a weak magnetic field. In the emission spectrum of pure strongly compressed Ge ⟨100⟩ one observes the emission lines of the EHL (L), of the free exciton (FE), and of the excitonic molecule (M), on which a weak magnetic field acts quite differently. Figure 3 shows the emission spectra of Ge ⟨100⟩ in magnetic fields 0, 0.6, and 2 T. The field 0.6 T was chosen because the stability of the EHL decreases in it and the L line drops out of the Ge ⟨100⟩ luminescence spectrum.[7] It can be seen from Fig. 3 that in a field $H = 0.6$ T the LP line drops out of the spectrum simultaneously with the L line. In the 2 T field, when the excitonic molecules become destabilized[9] and the stability of the EHL is restored,[7] the M line drops out of the emission spectrum, but the L and LP lines appear. These facts demonstrate incontrovertibly the hole origin of the LP line.

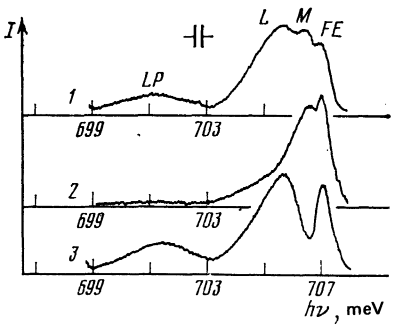

FIg. 3. Change of Ge $\langle 100\rangle$ emission spectra in a magnetic field (H): 1–0, 2–0.6, 3–2 T at $P = 470$ MPa and $T = 1.5$ K.

The position, the low intensity, and the behavior of the LP line in a weak magnetic field indicate that this line is also due to recoil that occurs when electron-hole pairs recombine into an EHL. As already noted above, for such processes it is extremely useful to compare the forbidden and allowed components of the spectrum. Figure 4 shows the forbidden and allowed components, shifted by a distance equal to the energy difference between the LA and TA phonons, as well as a plot of $I_{TA}/I_{LA}(h\nu)$. The increase by 2.5 times of the relative intensity of the LP line in the forbidden component compared with the allowed one and the constancy of the ratio $I_{TA}/I_{LA}(h\nu)$ in the region of the LP line (Fig. 4) demonstrate that the LP line is indeed due to recoil processes with participation of particles whose energy is practically independent of the quasimomentum. The foregoing peculiarities, as well as the spectral position of the LP line (which is shifted relative to the line towards the red by an energy ~ 4 meV close to the characteristic plasma frequency of an EHL with equilibrium density 7×10^{15} cm^{-3}, $\hbar\omega_p \approx 3.7$ meV), allow us to conclude that the LP line appears in the emission spectrum because of recombination in the EHL with participation of a plasmon. We emphasize that the LP line cannot be due to recoil processes in which the Aguer carrier is thrown over into the band that splits away as a result of the deformation.[6] In this approach, the energy spacing $\Delta h\nu$ in the spectrum between the lines L and LP should increase linearly with the strain in accord with the splitting of the energy bands, but the experimentally observed $\Delta h\nu(P)$ dependence, on the

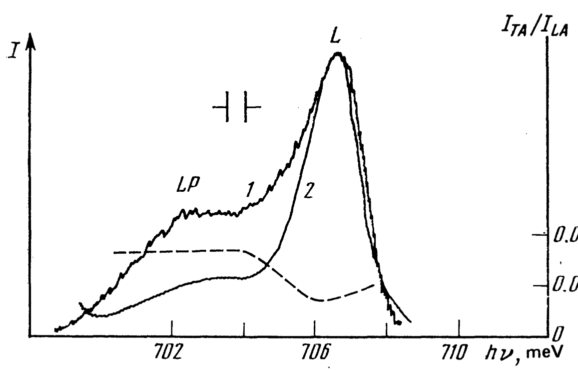

FIG. 4. Comparison of Ge $\langle 100\rangle$ emission spectra in the forbidden (1) and allowed (2) components of the spectrum. The TA spectrum component is shifted by the energy difference between the LA and TA phonons. The dashed curves is a plot of I_{TA}/I_{LA} vs the energy of the emitted photon.

contrary, saturates after first decreasing, and remains constant starting with $P = 350$ MPa up to $P = 500$ MPa (§4). An LP line with a somewhat smaller value of α was observed also in the emission spectrum of Ge $\langle 111\rangle$ at $P > 300$ MPa and $T < 2$ K.

§4. DEPENDENCE OF THE EQUILIBRIUM EHL DENSITY IN Ge$\langle 100\rangle$ AND Ge $\langle 111\rangle$ ON THE STRAIN

As noted above, in the case of the low density EHL produced in deformed Ge, the usual method of determining the EHL density, based on approximating the L-shape, is not reliable. The observed LP line corresponding to recombination of electron-hole pairs into an EHL with participation of a plasmon can therefore be used to obtain an independent estimate of n. In contrast to undeformed Ge, where the macroscopic properties are not anisotropic because of the cubic symmetry of the crystal, and where the plasma frequency is determined by the reduced optical mass (μ), in deformed Ge, where the crystal symmetry is lowered, the plasma frequency depends on the direction of propagation of the oscillations. Thus, in the Ge $\langle 111\rangle$ case the plasma frequency is determined by two masses: $\mu_\parallel = 0.039 m_0$ and $\mu_\perp = 0.050\, m_0$ along and across the compression direction.

A description of the LP line shape (in the case of allowed transitions) can be obtained by using the Fermi golden rule.

$$\frac{\hbar}{2\pi\tau(\nu)} = |M|^2 \int_0^\infty \int_0^\infty \int_0^\infty d\varepsilon_h D_e(\varepsilon_e) D_h(\varepsilon_h) D_p(\varepsilon_p) f_e(\varepsilon_e) f_h(\varepsilon_h)$$

$$\times f_p(\varepsilon_p)\,\delta(\varepsilon_e+\varepsilon_h+E_g-\varepsilon_p-\hbar\Omega-h\nu), \quad (3)$$

where E_g is the energy gap of the compressed Ge, $\hbar\Omega$ is the energy of the LA phonon, M is a transition-matrix element, and D_i and f_i ($i = e,h,p$) are the state densities and the distribution functions of the particles (electrons, holes, and plasmons). In the case of Ge $\langle 111\rangle$ the plasmon energy, neglecting the weak dependence on the quasimomentum, can be written in the form

$$\hbar\omega_p(\theta) = (\varepsilon_1^2 \sin^2\theta + \varepsilon_2^2 \cos^2\theta)^{1/2},$$

$$\varepsilon_1 = (4\pi e^2 n/k\mu_\perp)^{1/2}, \qquad \varepsilon_2 = (4\pi e^2 n/k\mu_\parallel)^{1/2}, \quad (4)$$

where $\mu_{\parallel,\perp}$ are the components of the tensor of the reduced effective mass of the electron and hole $(\mu_{ij})^{-1} = (m_{ij}^e)^{-1} + (m_{ij}^h)^{-1}$ and θ is the angle between the principal axis of the tensor μ_{ij} and the propagation direction of the plasma oscillation. From relations (4) and (3) we obtain for the LP-line shape

$$I(h\nu) \sim \int_{\varepsilon_1}^{\varepsilon_2} D_p(\varepsilon) f_p(\varepsilon) F(h\nu-\varepsilon)\, d\varepsilon, \quad (5)$$

where the density of the plasmon states is $D_p(\varepsilon) \sim \varepsilon / [(\varepsilon^2 - \varepsilon_1^2)(\varepsilon_2^2 - \varepsilon^2)]^{1/2}$, and

$$F(h\nu) = \int_0^{h\nu} D_e(\varepsilon) f_e(\varepsilon) D_h(h\nu-\varepsilon) f_h(h\nu-\varepsilon)\, d\varepsilon \quad (6)$$

is the L-line shape. The LP-line shape is thus a convolution of the line L with the plasmon-state density.

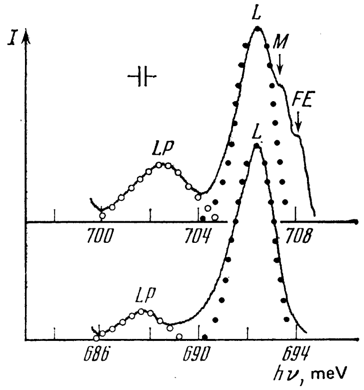

FIG. 5. Approximation of the shapes of the emission spectra of Ge ⟨100⟩ and Ge ⟨111⟩ by expressions (5) and (6).

The shapes of the lines L and LP, as well as their relative positions in the emission spectrum, are determined by only one adusting parameter, the EHL density, which is determined under these conditions with good accuracy. Figure 5 shows approximations of the shapes of the line L and LP by expressions (5) and (6), from which were obtained the equilibrium line densities $n = (10.5 \pm 0.5)\times 10^{15}$ cm^{-3} in Ge⟨111⟩ at $P = 440$ MPa and $n = (8.1 \pm 0.4)\times 10^{15}$ cm^{-3} in Ge ⟨100⟩ at $P = 480$ MPa. These values are in good agreement with the data of Ref. 7.

Uniaxial deformation, as already noted, lowers the state density in the bands, and this leads to a decrease of the EHL binding energy (φ) and of its equilibrium density. At pressures $P > 200$ MPa, when the band splitting exceeds the corresponding Fermi-carrier energies, the decrease of the state density, and hence of the values of φ and n, is due only to the decrease of the nonparabolicity of the valence band. The $n(P)$ dependence was analyzed theoretically in Ref. 10. The change of the EHL equilibrium density with increasing strain was determined experimentally, by the method described above, for Ge ⟨100⟩ in the interval 200 MPa $< P <$ 500 MPa, and for Ge ⟨111⟩ in the interval 300 MPa $< P <$ 450 MPa. Figure 6 shows the experimental and theoretical $n(P)$ dependences, which appear to be in satisfactory agreement.

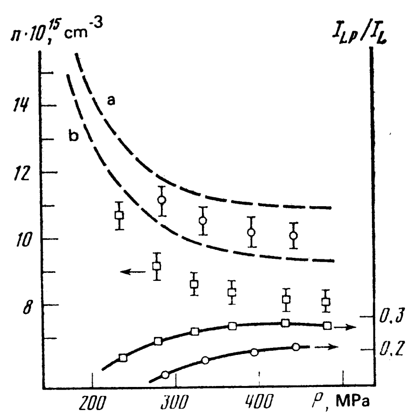

FIG. 6. Experimental dependences of the EHL density and of the relative intensity of the LP line on the strain in Ge ⟨100⟩ (□) and in Ge ⟨111⟩ (○), obtained at $T = 1.8$. The dashed curves show the $n(P)$ dependences calculated from the equations of Kirczenow and Singwi[10] for Ge ⟨111⟩ (a) and Ge ⟨100⟩ (b) at $T = 0$ K.

§5. DEPENDENCE OF THE RELATIVE INTENSITY OF THE LP LINE ON PRESSURE AND TEMPERATURE

As noted above, the relative intensity of the LP line depends on the strain, on the temperature, and on the focusing conditions. One of the characteristic features of the dependence of $I_{\rm LP}/I_L$ on the focusing conditions is the redistribution of the radiation intensity between the LP line corrresponding to the process of $e - h$ pair recombination into EHL with participation of a plasmon, on the one hand, and the red tail of the L line, which is due to recombination with participation of an Auger electron, on the other. The LP line appears here on account of the partial decrease of the radiation intensity on the red edge of the L line and the increase of the radiation in the spectrum region with still lower energy (see Fig. 2). We point out in this connection that such a transformation of a plasmon into an Auger electron (more accurately, into an $e + h$ pair), is known as Landau damping, according to which the plasmon as a quasiparticle gives up all its energy and quasimomentum to excitation of a separate carrier outside the Fermi sphere.[11] In this case, as noted in Ref. 12, since the wave vector of the plasmon in an electron-hole drop (EHD), in contrast to an unbounded medium, is a poor quantum number in view of the small size of the EHD, the quasimomentum conservation law is satisfied only accurate to h/d, where d is the EHD diameter. This circumstance increases greatly the role of Landau damping in EHD plasma oscillations. The theoretical estimate of this damping, obtained in Ref. 12, is $\gamma_L \approx e^2/d$. The dependence of the Landau damping on the EHD size casts light on the problem of the change of the relative intensity of the LP line as a function of the focusing conditions, since it is well known that these conditions determine the size of the EHD.[13]

In addition to the Landau-damping mechanism described above, there exists a damping mechanism based on electron-hole collisions that disrupt the coordinated motion of the individual carriers. Within the framework of this mechanism, the plasma-oscillation damping is described by the quantum-mechanical expression $\gamma_{eh} = AT^2 + B\omega^2$, which was experimentally verified in the case of EHD in

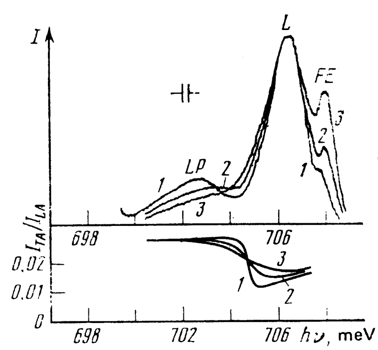

FIG. 7. Change of the emission spectra of Ge ⟨100⟩ at $P \sim 420$ MPa and $H = 1.9$ T with change of bath temperature (T_b): 1–1.5, 2–1.9, 3–2.1 K. In the lower part of the figure is shown the temperature dependence of the ratio $I_{TA}/I_{LA}(h\nu)$.

undeformed Ge.[14] According to this expression, the damping of the plasma oscillations should decrease when the crystal is deformed, for this decreases the equilibrium density of the EHD and the frequency of the plasma oscillations; it should also be decreased when the temperature is lowered.

Figure 6 shows a plot of I_{LP}/I_L vs the crystal deformation along the $\langle 111\rangle$ and $\langle 100\rangle$ directions at $T = 1.8$ K. It can be seen from the figure that at fixed temperature and fixed focusing the value of I_{LP}/I_L is determined by the equilibrium density of the EHD. The change of the emission spectra with increasing temperature in the presence of the magnetic field needed to destabilize the exitonic molecules is shown in Fig. 7. A characteristic feature is that the transformation of the emission spectrum with increasing temperature is similar to that described above: when the intensity of the LP line is decreased, the intensity of the red tail of the L line increases. This is evidence that raising the temperature increases the probability of processes in which the recoil particle is an Auger electron rather than a plasmon. The electron and plasmon dispersion laws differ substantially, and this is reflected in the variation of the $I_{TA}/I_{LA}(h\nu)$ dependence shown in Fig. 7.

We have thus succeeded in observing for the first time recombination radiation of an EHL with participation of a plasmon, and this has enabled us to obtain a reliable estimate of the equilibrium density of the EHL in strongly deformed Ge.

In conclusion, the author is deeply grateful to V. D. Kulakovskiĭ and V. B. Timofeev for helpful discussions and for interest in this work.

[1)]A deviation of 3–5° from the $\langle 100\rangle$ axis is necessary to lift the degeneracy in the conduction band.

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