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QUANTUM OPTICS

Interaction of Phase-Modulated Femtosecond Pulses with an Optically Dense Quasi-Resonant Medium of Rubidium Vapors

S. N. Bagaev[a], A. A. Preobrazhenskaya[b], N. A. Timofeev[b], A. A. Pastor[b], I. B. Mekhov[b, c, d], I. A. Chekhonin[b, *], P. Yu. Serdobintsev[b], V. S. Egorov[b], M. A. Chekhonin[b], and A. M. Mashko[b]

[a] *Institute of Laser Physics, Siberian Branch, Russian Academy of Sciences, Novosibirsk, 630090 Russia*
[b] *St. Petersburg State University, St. Petersburg, 199034 Russia*
[c] *University of Oxford, Oxford, United Kingdom*
[d] *University Paris-Saclay, Paris, France*
**e-mail: chekhonin@mail.ru*

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Abstract—For the first time, it is demonstrated that the magnitude and sign of the effect of "spectral condensation" of a laser pulse at the resonant-transition frequency of a dense medium can be controlled by changing the driving-pulse parameters (chirp, pulse width, and pulse amplitude). In the process of this, importantly, the driving-pulse energy and spectrum remain unchanged. Direct time-resolved measurements revealed an oscillatory character of the induced superradiance of rubidium vapors representing a long train of decaying short pulses. The width and repetition rate of the pulses in the train are determined by atomic density N_0 of the medium, while the width of an entire superradiance pulse (10 ps) is considerably larger than that of the driving laser pulse (50 fs).

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INTRODUCTION

The propagation of shaped ultrafast laser pulses in resonant media traditionally attracts much attention. The reason for this is the fact that the rate of multiphoton transitions for such pulses under nonlinear interaction can be substantially increased despite lower peak pulse power.

Propagation of frequency-chirped pulses in rubidium vapors in the adiabatic-following regime, along with formation of self-induced-transparency solitons, was investigated in [1, 2]. The authors of [3] demonstrated that the "absorption-line" spectrum under coherent regime of pulse propagation in an extended dense resonant medium reveals a characteristic fine oscillatory structure.

It was noted in [4] that maximizing nonlinear interaction of light with matter is generally assumed to be the primary means of laser-pulse compression for obtaining transform-limited ultrashort pulses (USPs). However, the authors demonstrated this not to be a universal principle.

On the contrary, it was demonstrated in [4] that the rate of resonant multiphoton transitions can be considerably increased, exceeding the level achievable by maximizing the peak pulse intensity only, by proper modification of the pulse spectrum and envelope (pulse shaping). As a proof of the principle, an experiment was conducted on observation of resonant two-photon absorption, wherein peak pulse intensity was decreased by a factor of 40 by selective suppression of some of the spectral bands. In so doing, the rate of quantum transitions was doubled. Furthermore, by properly choosing the spectral phase of the pulse, the authors were able to increase the rate of quantum transitions by a factor of 7.

In [5], the authors increased the peak population of the excited levels relative to that achievable with the help of transform-limited pulses by applying pulse-shaping methods to a broadband 100-fs pulse that was in resonance with a two-photon atomic transition. The authors also demonstrated that dispersion induced by an absorption line itself leads to analogous fast-oscillating transient processes in an excited state. These effects are applicable to any multiphoton resonant transition.

It is well known [6] that coherent propagation of a USP through a resonantly absorbing medium leads to pulse splitting and formation of a train the pulses the first of which represents the input pulse, while the suc-

cessive pulses in the train represent a response of the resonant-medium polarization (the 0π pulse).

It also known [7, 8] that such a regime of propagation leads to the effect of "spectral condensation" in which broadband input radiation of a femtosecond laser becomes concentrated near the resonant absorption lines at the output of an optically dense medium. It was demonstrated that this effect is related to excitation of a resonant polarization in the form of a nonadiabatic quantum jump, while emission of the medium represents an inversionless induced superradiance of an extended dense resonant medium. A similar effect is also observed in lasers with an intracavity absorption cell operating in the mode-locking regime [9] and in free space [10, 11].

In the present work, we present the results of studying coherent propagation of an USP through optically dense rubidium vapors under quasi-resonant conditions. Since such an interaction process was analyzed in earlier publications without taking into account the pulse chirp [7, 8], it would be of interest to find out how chirping of the input pulse affects the pulse of the medium superradiance. Here, we briefly present the results of both spectral and temporal measurements of the laser pulse at the output of the medium.

EXPERIMENTAL SETUP

The experiments were conducted with a Pulsar-10 femtosecond laser with the following parameters: pulse width of 50 fs, pulse energy up to 10 mJ, central wavelength of 790 nm, spectral width of the pulse of about 20 nm, light intensity in the beam of 0.1 TW/cm^2, and beam diameter of 5 mm. Phase modulation was introduced and controlled by means of an optical compressor formed by two diffraction gratings the distance between which could be varied. Rubidium vapors contained in a cell (10-cm length, 3-cm diameter) filled with a buffer gas (argon) were used as a resonant medium. To obtain required pressure of rubidium vapor, the cell was heated to the temperature of 200–400°C. Concentration of rubidium vapors was varied between 7.6×10^{14} and 1.7×10^{17} cm^{-3}.

The Pulsar-10 laser used in the experiments had an architecture of a chirped-pulse amplification (CPA) system and consisted of an oscillator generating 50-fs pulses, a stretcher for increasing the pulse width before amplification, an amplifier, and a compressor formed by two diffraction gratings.

The input pulse was chirped by changing the distance between the gratings by L to either side from the length optimal for pulse compression. The laser pulse was assumed to have a Gaussian shape with width Δt_0. The layout of the grating compressor is illustrated in Fig. 1.

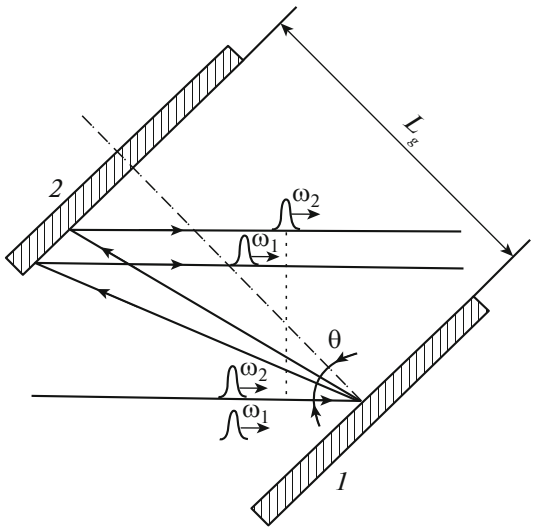

Fig. 1. The scheme of a grating compressor. *1* and *2* are identical gratings.

The shape of the output pulse was determined by group-delay dispersion G_d defined by the dependence

$$G_d(L) = 5250L \text{ (mm) [fs}^{-2}\text{]}$$

in our experiments. Here, L is the distance by which the grating distance was changed relative to the position corresponding to minimal output-pulse width and highest peak power.

The pulse at the output of the compressor had an amplitude and frequency (phase) modulation:

$$E(t) = A(t, G_d)\sin[(\omega_0 + \Delta\Omega(t, G_d))t].$$

Here, $A(t, G_d)$ is the slowly varying envelope of the Gaussian pulse, ω_0 is the central carrier frequency of the laser field, and $\Delta\Omega(t, G_d)$ is the deviation of the laser-field frequency. It was assumed that $\Delta\Omega(t)$ is a linear function of time.

In general, the chirp magnitude is given by

$$C(G_d) = d(\Delta\Omega)/dt.$$

Changing grating distance affects three parameters simultaneously: pulse amplitude $A(t, G_d)$, pulse width $\Delta t(G_d)$, and chirp $C(G_d)$. It is important to underscore that the laser-pulse energy and spectrum remain unchanged.

The dependences of pulse width $\Delta t(G_d)$ and chirp $C(G_d)$ on compressor detuning are illustrated in Figs. 2 and 3, respectively.

Figure 2 represents variation of the pulse width as a function of detuning L given by the expression

$$\Delta t^2 = (\Delta t_0^4 + 16(\ln 2)^2 G_d^2)/\Delta t_0^2,$$

where Δt_0 is the pulse width at zero compressor detuning ($L = 0$).

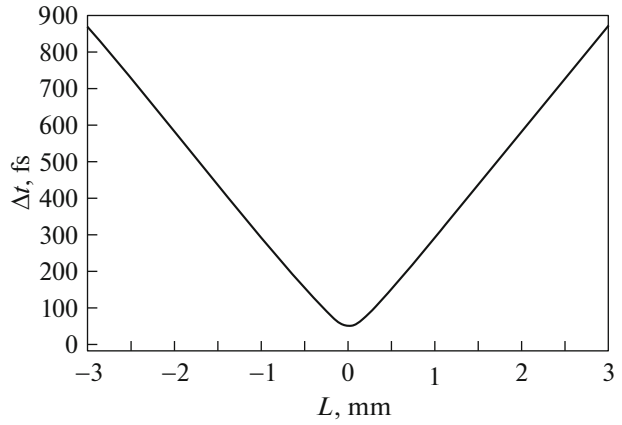

Fig. 2. Pulse width as a function of compressor detuning L.

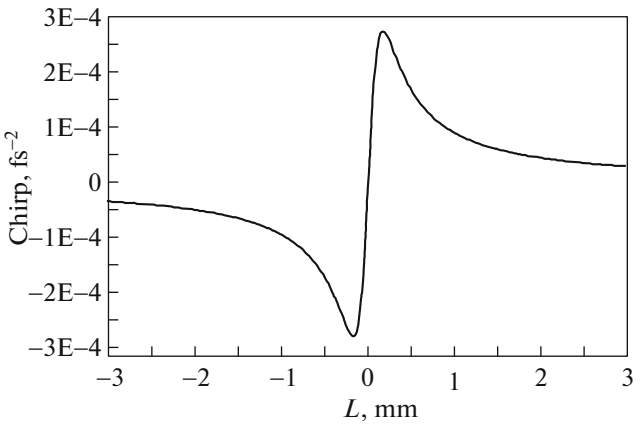

Fig. 3. Pulse chirp $C(G_d)$ as a function of compressor detuning L.

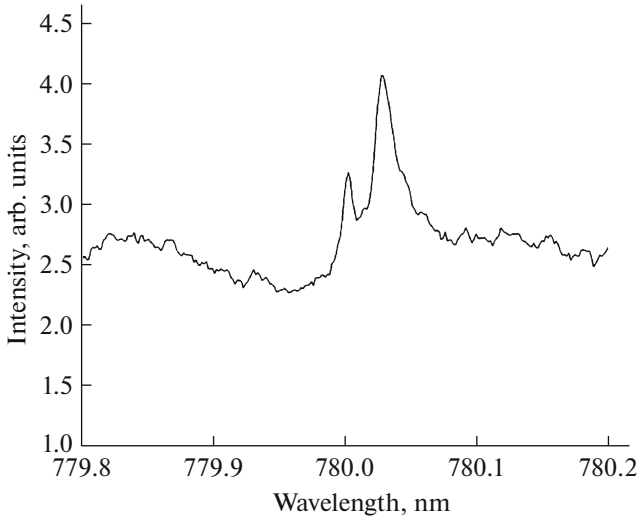

Fig. 4. The effect of "spectral condensation" in the vicinity of a resonant line of rubidium (wavelength 780 nm). $G_d = -2610$ fs^{-2}.

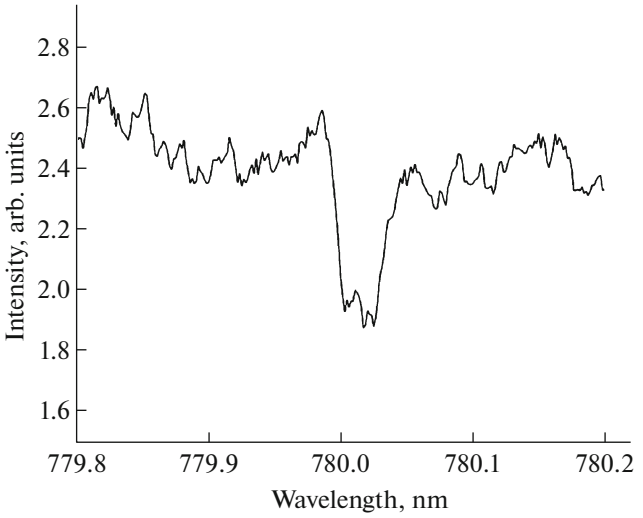

Fig. 5. The effect of "spectral anticondensation" in the vicinity of a resonant line of rubidium (wavelength 780 nm). $G_d = 2610$ fs^{-2}.

Figure 3 illustrates the dependence of chirp $C(G_d)$ on compressor detuning that is described by the following dependence:

$$C(G_d) = 8(\ln 2)^2 G_d/(\Delta t_0^4 + 16(\ln 2)^2 G_d^2).$$

Examples of "condensation" and "anticondensation" of the femtosecond-pulse spectrum after propagation through a cell with rubidium at different signs of the pulse chirp at resonant frequency of rubidium D_2 with a wavelength of 780 nm are presented in Figs. 4 and 5, respectively.

It is obvious that the effect of "spectral anticondensation" takes place upon changing the sign of the chirp, i.e., emission of the medium near the absorption line in the spectrum is suppressed.

The spectra of laser radiation after propagation through the cell with rubidium in the "spectral condensation" regime were obtained by using a high-resolution DFS-8 spectrograph with a 1200 gr/mm grating and dispersion of 0.4 nm/mm. The spectral width of the laser radiation was found to be 20 nm. A CCD camera with a 7-mm matrix (1000 pixels) was used for signal detection. As a result, we were able to obtain spectra only in the 2-nm interval, which prohibited us from obtaining an entire spectrum simultaneously. A panoramic spectrum was obtained by means of a USB-2000 spectrometer (Ocean Optics) with a spectral resolution of 0.5 nm. Such a spectrum is presented in Fig. 6.

The upper spectrogram represents the spectrum of laser radiation after propagation through the cell with

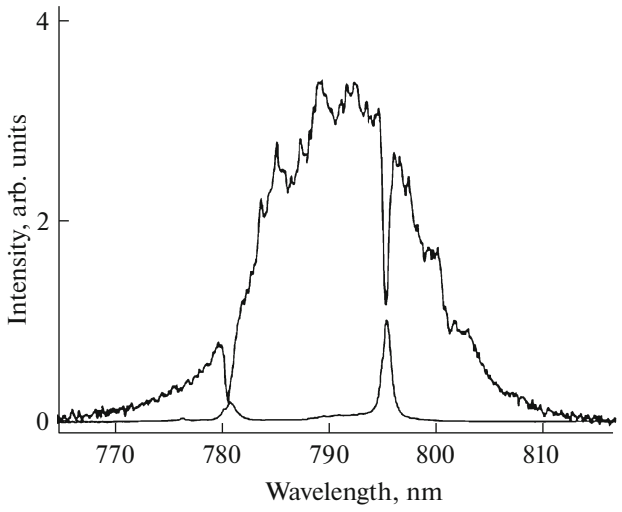

Fig. 6. A panoramic spectrum obtained with a USB2000 spectrometer (Ocean Optics).

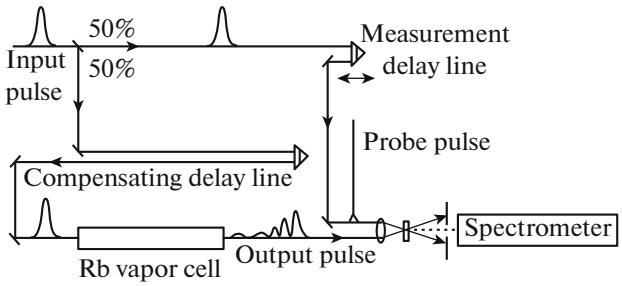

Fig. 7. Experimental setup. The main components are a femtosecond laser, a cell containing rubidium vapors, optical delay lines, and a USB2000 spectrometer (Ocean Optics).

rubidium under the conditions of incoherent propagation. To this end, the laser beam before the cell was passing through a Teflon diffuser. The spectrum reveals two absorption lines corresponding to the resonant doublet of rubidium. The lower spectrogram was obtained under the conditions of coherent propagation (without a diffuser). The spectrogram reveals the effect of spectral condensation in the vicinity of the two resonant lines of rubidium. It can be seen from the panoramic spectrum presented in Fig. 6 that the effect of condensation reveals itself at both the D_1 and the D_2 lines of the resonant doublet of rubidium.

We used rubidium vapors containing a natural mixture of isotopes with atomic weights of 85 and 87, respectively. However, isotopic splitting of rubidium lines D_1 and D_2 is much smaller than the observed width of the lines of spectral condensation (anti-condensation).

The effect of "spectral condensation" can be observed with two types of spectra at the resonant atomic-transition frequency. At low density N_0 of rubidium atoms, a narrow line of superradiance is observed at atomic-resonant frequency ω_{12}. The resonant-emission intensity increases with an increase in atomic concentration.

However, starting from a certain threshold value of N_0, the emission spectrum of the atomic ensemble splits into two satellites [7]. The magnitude of their frequency splitting is determined by the value of frequency splitting of polariton modes in the resonant medium and increases with an increase in N_0.

The obtained results show that the response time of the resonant polarization of the medium to an input pulse is very sensitive to the sign of the phase modulation (chirp). It was demonstrated in [8] that the time delay between them can lead to either a constructive interference of the input pulse with the response pulse of the medium polarization or to a destructive interference (upon changing the sign of the chirp).

This circumstance allows explaining the transition between two types of spectrum of the output radiation: "spectral condensation" and "spectral anticondensation." Hence, there exists a possibility of controlling the magnitude and sign of the effect of "spectral condensation" without changing the laser-pulse energy or spectrum.

DIRECT MEASUREMENTS OF TEMPORAL PARAMETERS OF THE PULSE PROPAGATED THROUGH AN OPTICALLY DENSE MEDIUM

It was suggested earlier in [7] that the effect of "spectral condensation" in rubidium vapors is related to the fact that a femtosecond laser pulse acting upon atoms of the medium induces a fast nonadiabatic transition (quantum jump) resulting in a quantum superposition of atomic states. A thus-prepared resonant medium has the form of an ensemble of phased dipoles the emission of which constructively interferes in the incident-pulse direction of propagation.

Such emission of the medium can be caused by a decay of collective free induction or an induced superradiance. Note that the appearance of this kind of superradiance does not require population inversion.

Depending on density N_0 of resonant atoms, the temporal dynamics of the superradiance pulse has two regimes: the regime of weak coupling between the field and the matter (emission of a single pulse with a monochromatic spectrum) and the regime of strong coupling (emission of an oscillatory pulse of superradiance in the form of an optical ringing characterized by a doublet form of its spectrum) [7].

In the present study, we carried out direct measurements of temporal characteristics of the femtosecond laser pulse propagated through an optically dense resonant medium of rubidium vapors. The measurements were conducted by using the method [6] of noncollinear second-harmonic generation, wherein

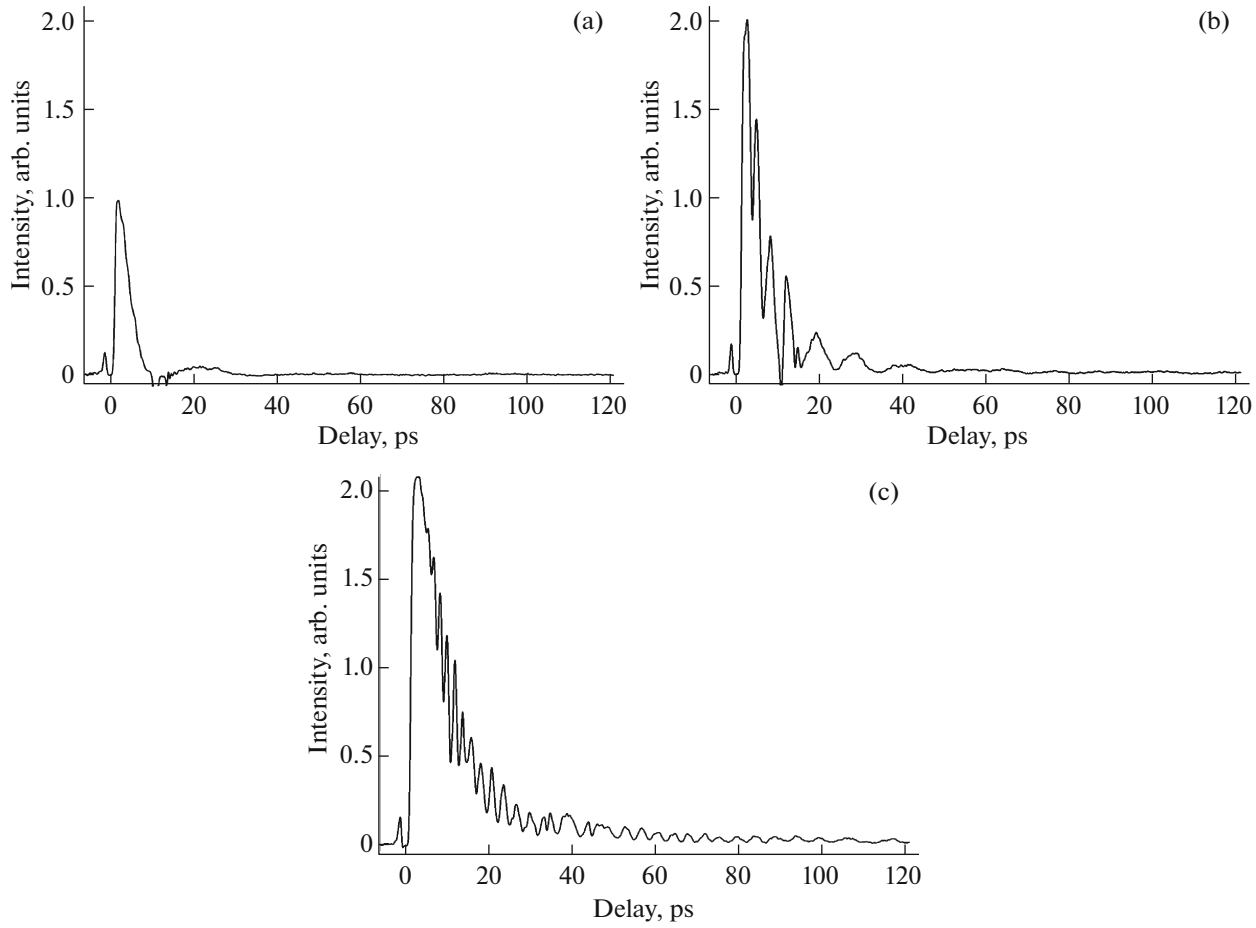

Fig. 8. Temporal dependence of the train of output pulses as a function of temperature of the cell filled with rubidium vapors. $T =$ (a) 200, (b) 300, and (c) 400°C. Pulse chirp is $C(G_d) = 0$.

the input pulse was mixed with a single probe pulse in a potassium dihydrogen phosphate crystal at different delays between the two pulses. The experimental arrangement is depicted in Fig. 7.

When conducting the time-resolved measurements, the grating compressor was tuned so that the pulse had no chirp: $C(G_d) = 0$ ($L = 0$).

Density of atoms N_0 under our experimental conditions corresponded to the strong-coupling regime (an oscillatory pulse of superradiance in the form of an optical ringing).

The obtained results show that the oscillatory superradiance of rubidium vapors had a duration of $\approx$10 ps for a driving laser pulse of width $\Delta t = 50$ fs. The duration of pulses in the train and their period were fully determined by atomic density N_0 of the medium. The results of the time-resolved measurements of the train of output pulses as a function of temperature of the cell containing rubidium atoms are presented in Fig. 8.

CONCLUSIONS

In the present work, we demonstrated for the first time that the magnitude and sign of the effect of "spectral condensation" of a laser pulse at the frequency of a resonant transition of a dense resonant medium can be controlled by changing temporal parameters of the driving pulse (chirp, pulse width, and pulse amplitude). Importantly, the driving-pulse energy and spectrum remain unchanged.

We believe that the change of sign of the effect is caused by variation of the delay between the driving pulse and the pulse of superradiance of the induced resonant polarization of the medium which is regulated by chirp value. The magnitude of the time delay is the main reason for transition from constructive to destructive interference of the pulses emerging from a dense medium.

Direct time-resolved measurements revealed that the pulse of an induced superradiance of rubidium vapors has an oscillatory character and represents a decaying train of short pulses. The width and period of the pulses in the train are determined by density N_0 of

atoms in the medium, while the duration of an entire pulse of superradiance considerably exceeds that of the driving laser pulse.

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