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Spectroscopic measurement of the softness of ultracold atomic collisions

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The softness of elastic atomic collisions, defined as the average number of collisions each atom undergoes until its energy decorrelates significantly, can have a considerable effect on the decay dynamics of atomic coherence. In this paper we combine two spectroscopic methods to measure these dynamics and obtain the collisional softness of ultracold atoms in an optical trap: Ramsey spectroscopy to measure the energy decorrelation rate and echo spectroscopy to measure the collision rate. We obtain a value of 2.5(3) for the collisional softness, in good agreement with previously reported numerical molecular-dynamics simulations. This fundamental quantity is used to determine the *s*-wave scattering lengths of different atoms but has not been directly measured. We further show that the decay dynamics of the revival amplitudes in the echo experiment has a transition in its functional decay. The transition time is related to the softness of the collisions and provides yet another way to approximate it. These conclusions are supported by Monte Carlo simulations of the full echo dynamics. The methods presented here can allow measurement of a generalized softness parameter for other two-level quantum systems with discrete spectral fluctuations.

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I. INTRODUCTION

Elastic collisions are of great importance in atomic physics from both a theoretical and a practical perspective. They are relevant for atomic clocks, metrology, quantum information, evaporative cooling, atom-ion hybrid systems, and more [1–6]. Collisions may also have a significant effect on the coherence properties of an ensemble of atoms, providing either elongation [7–16] or shortening [17,18] of the atomic coherence time.

Considering a rapid collisional process compared to other dynamical time scales,¹ there exist two extremities for a colliding atom in the center-of-mass frame of the interacting ensemble: hard collisions, in which the energy of the atom is completely randomized after a single collision, and soft collisions, in which the atomic energy remains almost unchanged after each collision [14]. We therefore define the collisional softness parameter s as the number of times an atom has to collide in order for the correlation between its initial and final energies to drop to $1/e^2$. The collisional softness of hard collisions is one, since the energy correlation drops to zero after a single collision. Collisions are considered soft if their softness parameter is much larger than unity. Even though the s-wave collisional process considered here is itself of universal nature, the softness of the collisions can be affected by the confining potential. This can be intuitively understood by considering that only the kinetic energy changes due to

a collision whereas the potential energy does not, carrying a memory of the total energy prior to the collision.

More formally, an ensemble of colliding trapped thermal atoms has two relevant characteristic rates. First, the atomic collisions, treated as Poisson process energy-randomization events, occur at an average collision rate Γ_{coll} . Second, the single-atom temporal energy autocorrelation function, averaged over the atomic ensemble, decays exponentially with an energy decorrelation rate Γ . The collisional softness is then defined as

$$s = \Gamma_{\rm coll} / \Gamma. \tag{1}$$

Collisions with s = 1 are hard and collisions with $s \gg 1$ are soft. The softness is the number of collisions required for thermalization in a perturbed trap, having immediate repercussions on the physics of evaporative cooling [19,20].

The softness of *s*-wave elastic collisions of ultracold bosonic atoms trapped in a harmonic potential and far from a Feshbach resonance was evaluated using molecular-dynamics simulations and found to be 2.5 [19–26]. This value of the softness has been used to determine the elastic collision cross sections of different atoms [19,21,22,25], but has not been measured directly.

In this paper we present a direct spectroscopic measurement of the softness of ultracold atomic collisions. We do so using a combination of two methods (Ramsey and echo [27]) in two opposite regimes of low and high collision rates. First we show that the coherence of an atomic ensemble in an echo experiment at low density asymptotically depends only on Γ_{coll} . We then show that in a high-density Ramsey experiment the decay is independent of the collision rate and can be fitted to reveal the energy decorrelation rate Γ . By combining these two measurements, we are able to quantitatively extract the *s*-wave collisional softness of cold ⁸⁷Rb atoms in an optical dipole trap. We obtain good agreement with previously reported theoretical results from molecular-dynamics simulations, validating our method and laying the foundation for its application in measuring the softness of other collisional processes.

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¹The collision time can be estimated by the range of atomic interaction divided by the relative velocity of the colliding atoms. For dilute ultracold atoms and excluding mean-field interactions, the collision time is on the nanosecond scale, much shorter than the mean time between collisions and the oscillation time in the trap ($\gtrsim 10$ ms and $\gtrsim 1$ ms, respectively for our experiment).

²This definition of *s* is related to the strength parameter of velocity changing collisions α , defined in [9], by $s = \frac{1}{1-\alpha^2}$.

We further show that the coherence decay in an echo experiment is qualitatively different for short and long times [8,9]. This can be used to approximate the softness by combining a Ramsey measurement and an echo measurement in a single, intermediate-density regime. These methods may allow measurements of a softness parameter for other two-level quantum systems that have discrete energy fluctuations [7,28–31].

II. SPECTROSCOPIC SIGNATURES OF COLLISIONAL SOFTNESS

⁸⁷Rb atoms trapped in an optical dipole trap experience a differential ac Stark shift imposed by the different detuning of the trapping laser from their two ground-state hyperfine levels. If the mean time between atomic collisions is larger than the oscillation period in the trap, the fast oscillations can be averaged. The rate of the phase accumulated by the wave function, determined by the detuning, then depends on the average energy [32]. Effectively this creates a stationary inhomogeneous broadening of the spectrum, decreasing the coherence time of the ensemble.

Due to this effect, the dynamics of the hyperfine coherence in a Ramsey $(\pi/2 - \pi/2)$ experiment with no collisions is given by

$$C_{\rm R}(t) = [1 + 0.95(t/\tau)^2]^{-3/2}$$
(2)

for an ensemble of two-level atoms in thermal equilibrium in an optical harmonic potential [32]. The bare Ramsey time is given by $\tau \approx 2\hbar/\eta k_B T$. Here *T* is the temperature of the cloud and η is the ratio between the hyperfine splitting and the detuning of the trapping laser.³ In an echo experiment $(\pi/2 - \pi - \pi/2)$, where the echo pulse is given at time t_{π} , the echo coherence $C_{\rm E}(2t_{\pi}) \equiv C(t = 2t_{\pi})$ fully revives in the absence of elastic atomic collisions due to the stationarity of the trap perturbation.⁴

Factoring in the effect of elastic atomic collisions, the Ramsey and echo coherences have a complicated behavior [11]. However, they both have some useful, simple limits. For high density n_{high} , the spectrum is collisionally narrowed, resulting in an elongated Ramsey coherence, which can be approximated by the generalized Gumbel function [15]

$$C_{\rm R}(t) \sim \exp\left[-\frac{2.86}{\Gamma^2 \tau^2}(e^{-\Gamma t} + \Gamma t - 1)\right],$$
 (3)

dependent only on τ and the energy decorrelation rate Γ and not on the collision rate Γ_{coll} .

In the opposite regime of low density n_{low} , the asymptotic long-time echo coherence behaves as

$$C_{\rm E}(2t_{\pi}) \sim \exp(-2\Gamma_{\rm coll}t_{\pi}). \tag{4}$$

In this regime the coherence depends solely on the collision rate Γ_{coll} and not on the energy decorrelation rate Γ . This is due to the fact that every collision, no matter how soft, causes a finite deflection in the atomic trajectory. As $t \to \infty$ this



FIG. 1. Comparison between echo revival in the regimes of (a) low collision rate ($\Gamma_{coll}\tau \approx 0.15$) and (b) high collision rate ($\Gamma_{coll}\tau \approx 10$). The Ramsey signal without the application of the echo (thin blue line) is compared to the echo signal after the application of the pulse (thick red line). Black dashed lines represent the times of the application of the π pulses. The envelope of the obtained Ramsey fringes is an indication of the atomic coherence. (a) At low collision rates the amplitude of the revival is high. The value of the coherence at the peak of the revival $C(t = 2t_{\pi})$ is defined as the echo coherence $C_{E}(2t_{\pi})$. (b) At high collision rates the echo pulse essentially has no effect on the coherence. The vertical axis represents the fraction of atoms in the upper hyperfine state after the last Raman pulse.

deflection will fully decohere the atom. This implies that the coherence in this long-time regime is nothing but the fraction of atoms that did not collide.

Figure 1 illustrates the role of collisions in an echo experiment. It presents the normalized upper hyperfine state population as a function of the time between the Ramsey pulses. In Fig. 1(a) the measurement is performed with very low collision rate ($\Gamma_{coll} \tau \approx 0.15$).⁵ The coherence decays fast and the echo revival amplitude is significant. On the other hand, in Fig. 1(b) the collision rate is much higher ($\Gamma_{coll} \tau \approx 10$). The Ramsey decay is slower (partially due to collisional narrowing) and the echo revival is negligible, manifesting the failure of the echo due to atomic collisions.

Both the collision rate and the energy decorrelation rate are proportional to the multiplication of the atomic density *n* with the average velocity $v_{rel} \sim \sqrt{T}$: $\Gamma_{coll} = n\sigma v_{rel}$. Therefore,

³For ⁸⁷Rb and a YAG 1064-nm trapping laser $\eta \approx 7 \times 10^{-5}$.

⁴For strong trap perturbations and for chaotic traps this may not be the case [33,34].

⁵Low and high collision rates are defined, throughout this paper, with respect to the bare Ramsey time τ . In both regimes the collision rate is much smaller than the trapping frequencies.

knowing the ratios $n_{\text{high}}/n_{\text{low}}$ and $T_{\text{high}}/T_{\text{low}}$ at the two different experimental conditions allows for the normalization of the collision rate, measured at low density, and the energy decorrelation rate, measured at high density, and extraction of the collisional softness

$$s = \left(\frac{n_{\text{high}}}{n_{\text{low}}} \sqrt{\frac{T_{\text{high}}}{T_{\text{low}}}}\right) \frac{\Gamma_{\text{coll}}^{\text{low}}}{\Gamma^{\text{high}}}.$$
 (5)

We note that in the intermediate regime $\Gamma \tau \approx 1$ it was shown theoretically that the spectrum of an ensemble of colliding atoms depends weakly on the softness [14]. It was further suggested that it may be possible to distinguish between the two extreme cases of hard and soft collisions, by measuring a Dicke narrowed spectrum in a Ramsey experiment. Practically, this task turns out to be challenging. Small uncertainties in the experimental conditions, such as the collision rate and the inhomogeneous broadening of the spectrum, may cause an incorrect model to fit well to the experimental data [10,11].

III. MEASURING THE COLLISIONAL SOFTNESS

Our apparatus is described in detail in [14]. Briefly, the experiment consists of ⁸⁷Rb atoms trapped in a 1064-nm far-detuned crossed-beam optical dipole trap. The atoms are evaporatively cooled down to two distinct regimes: high density with $n_{\text{high}} = 3.5(2) \times 10^{12} \text{ cm}^{-3}$ and $T_{\text{high}} = 0.56(2) \,\mu\text{K}$ and low density with $n_{\text{low}} = 3.6(3) \times 10^{11} \text{ cm}^{-3}$ and $T_{\text{low}} = 6.8(3) \,\mu\text{K}$.⁶ Measurement of the total number of atoms, and hence the peak density, is susceptible to common systematic errors and obtaining an exact value for it is challenging [35]. However, as our method relies only on the knowledge of the ratio between densities [Eq. (5)], systematic errors are common-mode rejected. All errors stated throughout the paper represent a 1 σ confidence level.

The coherence is measured between the first-order Zeeman insensitive hyperfine $|1\rangle \equiv |F = 1, m_F = 0\rangle$ and $|2\rangle \equiv |F = 2, m_F = 0\rangle$ states of the 5²S_{1/2} manifold. The atoms are prepared by optical pumping and microwave transitions in state $|1\rangle$. We then use a microwave ~6.8-GHz control to perform Ramsey ($\pi/2$ - $\pi/2$) or phase-scanned echo ($\pi/2$ - π - $\pi/2$) manipulations on the atoms. At the end of each experiment we use a state-selective fluorescence-detection scheme to evaluate the fraction of atoms at state $|2\rangle$. The coherence is defined as the normalized amplitude of the fringes of the Ramsey and echo data.

We measure the collisional softness using Eq. (5), by first obtaining Γ_{coll}^{low} from an echo measurement in the low-density regime by fitting the asymptotic decay described by Eq. (4) and then obtaining Γ^{high} , given by Eq. (3), from a Ramsey measurement in the opposite regime. Focusing first on the low-density regime, the resulting echo coherence and an additional Ramsey measurement at the same experimental conditions are



FIG. 2. Ramsey and echo experiments. (a) Atomic coherence as a function of time in Ramsey (blue circles) and echo (red squares) experiments at low density. The decay of the Ramsey signal yields a coherence time of $\tau_{low} = 37(1)$ ms. (b) Linear fit to the tail of the echo decay, in logarithmic scale, gives $\Gamma_{coll}^{low} = 9.4(3) \text{ s}^{-1}$. Crosses are short-time data points excluded from the fit. Solid lines represent the fitted functions (see the text). Time in the echo experiment corresponds to $2t_{\pi}$. (c) Ramsey measurement of the energy decorrelation rate Γ^{high} at high density. The measured coherence is fit to a Gumbel function [Eq. (3)] with the energy decorrelation rate as a fitting parameter. This yields $\Gamma^{high} = 10.6(1) \text{ s}^{-1}$.

presented in Fig. 2(a). The echo decay time is indeed much longer than that of the Ramsey experiment (by about a factor of 4). The extracted low-density bare Ramsey time is $\tau_{low} = 37(1)$ ms, compared to 32(1) ms obtained directly from the measured temperature. The echo measurement exhibits a long-time linear decay on a semilogarithmic scale [Fig. 2(b)], confirming the expected exponential decay of Eq. (4). The slope, excluding short times, gives a collision rate of $\Gamma_{coll}^{low} = 9.4(3) \text{ s}^{-1}$. Next we obtain the energy decorrelation rate Γ^{high} from the collisional narrowing of a high-density Ramsey measurement. The atomic coherence is shown in Fig. 2(c). From the measured temperature, we expect $\tau_{high} = 390(15)$ ms. We use this value as a fixed parameter and fit the coherence data to Eq. (3), extracting $\Gamma^{high} = 10.6(1) \text{ s}^{-1}$. The softness is then calculated using Eq. (5) to be s = 2.5(3),⁷ in excellent agreement with molecular-dynamics simulations [20].

We perform Monte Carlo simulations to study the effect of the softness of the collisions on the full dynamics of the

⁶The temperature is measured using time of flight and the peak atomic density using $n = \omega_x \omega_y \omega_z N (\frac{m}{2\pi k_B T})^{3/2}$, where ω_i are the trap frequencies, *N* is the total number of atoms, *m* is the atomic mass, k_B is the Boltzmann constant, and *T* is the measured temperature.

⁷This value is correct for the harmonic trapping potential that describes well our crossed Gaussian beam optical potential. For other trap shapes and energy distributions it may vary, e.g., for a flat box potential s = 1.5 [20].



FIG. 3. Echo experimental results (circles) compared to Monte Carlo simulations for atoms with an energy distribution corresponding to a 3D harmonic potential with hard (s = 1) (dashed purple line), soft (s = 10) (dash-dotted green line), and moderate (s = 2.5) (solid red line) collisions. The simulation uses the calculated bare Ramsey time of $\tau_{\text{low}} = 32$ ms and a constant energy decorrelation rate of the measured $\Gamma^{\text{high}} = 10.6 \text{ s}^{-1}$, rescaled using Eq. (5). Insets illustrate the energy of a sample atom as a function of time for hard (s = 1) (purple, top right) and soft (s = 10) (green, bottom left) collisions, with the same energy decorrelation rate Γ .

echo decay. The simulation calculates the ensemble coherence of 2×10^4 two-level atoms with the energy distribution corresponding to a three-dimensional (3D) harmonic potential [32] as a function of time. The collision rate Γ_{coll} is drawn from a Poisson distribution and the collisional softness s is generated by introducing controlled correlations between the energy jumps of successive collisional events using the Cholesky decomposition method of the required correlation matrix [36]. Typical energy trajectories for s = 1 and s = 10 are illustrated in the insets of Fig. 3. A comparison between the experimental data and simulation results is presented in Fig. 3 for the echo experiment with hard (s = 1), soft (s = 10), and moderate (s = 2.5) collisions, using the measured energy decorrelation rate, rescaled using Eq. (5), $\Gamma^{\text{low}} = (\frac{n_{\text{high}}}{n_{\text{low}}})^{-1} \Gamma^{\text{high}} =$ 3.76 s⁻¹. The simulation of the s = 1 and s = 10 collisions clearly disagrees with the data, whereas the s = 2.5 collision agrees best with the experimental data for all times and with no fit parameters.

IV. MEASURING THE SOFTNESS USING A TRANSITION IN THE FUNCTIONAL DECAY OF THE ECHO DYNAMICS

In the low-density regime of $\Gamma \tau \ll 1$, valuable information can be extracted by observing the entire dynamics of the decay of the echo coherence. Equation (4) gives the long-time limit of the coherence $C_{\rm E}(2t_{\pi}) \sim \exp(-2\Gamma_{\rm coll}t_{\pi})$, depending solely on the collision rate $\Gamma_{\rm coll}$. The short-time limit, however, depends on the energy decorrelation rate Γ and the bare Ramsey time τ and is given by [17]

$$C_{\rm E}(2t_{\pi}) \sim \exp\left[-\frac{\Gamma(2t_{\pi})^3}{6\tau^2}\right].$$
 (6)



FIG. 4. Echo measurement, with $\Gamma_{coll} t_{tr} \approx 1$, showing a transition of α . (a) Data are fit to the function given in Eq. (7), yielding $2t_{tr} \approx 205$ ms [dashed vertical lines in (a)–(c)], using $\Gamma_{coll} = 5.1(3) \text{ s}^{-1}$ obtained from (c) as a set parameter. The inset shows a Ramsey experiment performed under the same experimental conditions, fitted (solid line) to Eq. (2), yielding a Ramsey time $\tau = 76(3)$ ms. (b) Same data with the time axis rescaled to $(2t_{\pi})^3$, showing the transition clearly on a semilogarithmic scale. The linear behavior at short times (the solid line represents a linear fit) indicates the $e^{-(2t_{\pi})^3}$ dependence. (c) Same data on a semilogarithmic scale as a function of $2t_{\pi}$. The linear dependence at long times (the solid line represents a linear fit) indicates the $e^{-2t_{\pi}}$ decay.

In both limits the echo coherence decays as $C_{\rm E}(2t_{\pi}) \sim \exp[-\beta(2t_{\pi})^{\alpha}]$, with $(\alpha,\beta) = (1,\Gamma_{\rm coll})$ for long times or $(\alpha,\beta) = (3,\Gamma/6\tau^2)$ for short times. Defining $t_{\rm tr} \equiv \tau/\sqrt{s}$, a transition time between the two regimes, an interpolating function can be written to describe the entire dynamics, which is the accurate solution in the limit of soft collisions [9]:

$$C_{\rm E}(2t_{\pi}) \sim \exp\left\{-2\Gamma_{\rm coll}t_{\pi}\left[1-\sqrt{\frac{\pi}{2}}\frac{t_{\rm tr}}{2t_{\pi}}\mathrm{erf}\left(\frac{\sqrt{2}t_{\pi}}{t_{\rm tr}}\right)\right]\right\}.$$
 (7)

We measure the echo coherence decay with a moderate collision rate ($\Gamma_{coll}t_{tr} \approx 1$). The resultant coherence is summarized in Fig. 4. The transition of α is evident from the fit to Eq. (7). An indication for the $\alpha = 3$ decay is shown in Fig. 4(b), where we plot the echo revival amplitudes on a logarithmic scale against $(2t_{\pi})^3$. Figure 4(c) shows the same data on a semilogarithmic scale as a function of $2t_{\pi}$. Here the linear dependence at long times indicates the $\alpha = 1$ decay. A similar transition was observed for warm molecular gases in the limit of soft collisions [8].

More quantitatively, extracting t_{tr} from the echo decay using Eq. (7) and τ from the Ramsey decay under the same experimental conditions, we can evaluate the softness by the



FIG. 5. Numerical investigation of the effectiveness of using Eq. (7) as a fitting function for extracting the softness for a Gaussian energy distribution (closed symbols) and the energy distribution corresponding to a 3D harmonic potential (open symbols). The simulation calculates, for a predetermined input softness, a Ramsey decay curve as well as a full echo coherence decay curve. To approximate the obtained softness, we simultaneously fit the Ramsey decay to Eq. (3) and the echo decay to Eq. (7) with three fitting parameters: Γ , τ , and *s*. The black dotted line is where the obtained softness is equal to the real one.

definition of the transition time $s = (t_{tr}/\tau)^2$. Fitting the echo decay of Eq. (7) to the data of Fig. 4, using $\Gamma_{coll} = 5.1(3) s^{-1}$ obtained from fitting the long-time decay of Fig. 4(c), we get $2t_{tr} = 205(17)$ ms.⁸ This, in addition to a Ramsey experiment under the same experimental conditions [inset of Fig. 4(a)] that gives $\tau = 76(3)$ ms, yields $s = (t_{tr}/\tau)^2 = 1.8(3)$. The agreement with the theoretical value of 2.5 is not as good as for the measurement described in the preceding section.

We investigate the use of the interpolation function of Eq. (7) as a fitting function for extracting the softness using the Monte Carlo simulations described previously. We find that for a Gaussian energy distribution the method is quite precise. Figure 5 presents a summary of the softness obtained for a Gaussian energy distribution (closed symbols) by fitting Eq. (7) to the numerically simulated decay of coherence as a function of the input softness for different values of $\Gamma \tau$. For most cases, the output softness is very close to the input softness. The relative error is less than 10% as long as $\Gamma_{coll} t_{tr}$ is within the range $0.1 < \Gamma_{coll} t_{tr} < 10$. Outside this range, the value of the coherence at the transition time $C_{\rm E}(2t_{\rm tr}) \approx \exp(-2\Gamma_{coll} t_{\rm tr})$ is either too high or too low for the fitting procedure to accurately extract the transition time.

⁸To evaluate the error $2\Delta t_{\rm tr}$ we use the value $\Gamma_{\rm coll} \pm \Delta \Gamma_{\rm coll}$ as a fitting parameter. The upper 1σ confidence bound on $t_{\rm tr}$ is that which is obtained for $\Gamma_{\rm coll} + \Delta \Gamma_{\rm coll}$ and the lower bound is that which is obtained for $\Gamma_{\rm coll} - \Delta \Gamma_{\rm coll}$.

For the case of an energy distribution of a 3D harmonic trap similar to the one we have in the experiment the situation is different (Fig. 5, open symbols). The approximate solutions for the Ramsey [Eq. (3)] and echo [Eq. (7)] coherence fit only qualitatively, yielding significant errors similar to the ones observed in the experiment.

V. SUMMARY AND OUTLOOK

We have measured the softness of ultracold elastic atomic collisions using a combination of two spectroscopic methods: a measurement of the energy decorrelation rate obtained from the collisional narrowing of a Ramsey experiment with a high collision rate and a direct measurement of the collision rate obtained from an echo experiment with a low collision rate. The obtained collisional softness is 2.5(3), in excellent agreement with the value previously obtained by molecular-dynamics simulations. We have also demonstrated a transition in the functional decay of the echo coherence, from $\exp(-t^3)$ at short times to $\exp(-t)$ at long times [8]. This transition occurs at a softness-dependent time $t_{\rm tr} = \tau/\sqrt{s}$. We show that this transition in the functional decay can be used to approximate the softness at a single intermediate-density regime.

Our results validate the spectroscopic method, allowing for its use in measuring the softness of other, nontrivial, collisional processes such as extensions to higher temperature involving the inclusion of more partial waves in the scattering process, fermionic collisions and thermalization [37], interspecies hybrid collisions [38,39], and atom-ion collisions [5]. It can also be useful as a tool in simulating the efficiency of evaporative cooling [20], the investigation of high-density atom interferometers [40], and slow and stored light [11]. Our methods may allow measurements of a generalized softness parameter for other two-level quantum systems with discrete spectral jumps [28–31].

Our work can be extended to warm vapor systems where the bare Ramsey time is dominated by Doppler broadening and the presence of a buffer gas induces collisional narrowing [7]. As in our system, the effect of collisional softness on the Ramsey signal is in itself very small and challenging to detect [10,11]. Combining Raman Rabi, Ramsey, and echo spectroscopy at high- and low-collisional-rate regimes can provide an accurate measure of the collisional softness for different buffer gases. In this case, it is possible to change $\Gamma_{coll}\tau$ by simply altering the angle between the Raman beams without actively changing the density [11].

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