

Intensity Correlations and Mesoscopic Fluctuations of Diffusing Photons in Cold Atoms

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We study the angular correlation function of speckle patterns that result from multiple scattering of photons by cold atomic clouds. We show that this correlation function becomes larger than the value given by Rayleigh law for classical scatterers. These large intensity fluctuations constitute a new mesoscopic effect specific to atom-photon interactions, that could not be observed in other systems such as weakly disordered metals. We provide a complete description of this behavior and expressions that allow for a quantitative comparison with experiments.

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A wave propagating in a random medium undergoes multiple scattering and the intensity pattern resulting from interferences of the scattered waves with each other is known as a speckle pattern. The angular and time dependent properties of these patterns have been extensively studied [1–3]. They exhibit coherent mesoscopic effects and provide a sensitive probe to scattering properties of diffusive media. Quasiresonant elastic scattering of photons by cold atomic gases represents in this context an important issue, since it provides a new tool to study properties of cold atomic gases such as atomic dynamics. Photon propagation in atomic gases differs from the case of electrons in disordered metals [4] or of electromagnetic waves in suspensions of classical scatterers, due to the existence of atomic internal degrees of freedom coupled to the photon polarization. Some effects of a Zeeman degeneracy on the coherent backscattering [2,5] have been recently investigated in the weak scattering limit [6] in terms of phase coherence times [7].

The purpose of this Letter is to study the static angular correlation function of photons performing coherent multiple scattering in a cold atomic gas. The photon intensity correlation function between angular scattering channels is defined using the transmission coefficient T_{ab} by

$$C_{aba'b'} = \frac{\overline{\delta T_{ab} \delta T_{a'b'}}}{\overline{T_{ab}} \overline{T_{a'b'}}}. \quad (1)$$

Here, the overbar denotes a configuration average over both the position of atoms and their internal degrees of freedom (see below) and $\delta T_{ab} \equiv T_{ab} - \overline{T_{ab}}$. For classical scatterers, intensity fluctuations obey the Rayleigh law $C_{abab} = 1$. In the presence of a Zeeman degeneracy, angular correlations of speckle patterns and intensity fluctuations become larger than 1. This is a new and genuine mesoscopic effect specific to multiple scattering of photons by atoms.

Atoms are modeled as degenerate two-level systems denoted by $|j_g m_g\rangle$ for the ground state and $|j_e m_e\rangle$ for the excited state, where j is the total angular momentum and m

is its projection on a quantization axis. The levels are degenerate with $|m_g| \leq j_g$ and $|m_e| \leq j_e$.

We refer to the following possible experimental setup (Fig. 1). A light pulse is incident along a direction \hat{s}_a onto a dense enough atomic gas confined in a slab geometry. This pulse is detected along a direction \hat{s}_b after being multiply scattered (ab channel). A time τ later, a second pulse that corresponds to the $a'b'$ channel, is detected. We assume that the time τ is short enough so that the atoms stay at rest between the two pulses. The same measurement is repeated after a time $T \gg \tau$, during which the scatterers move. The averaging over spatial disorder results from this motion. This is a Young-like experiment. Thus, although a pulse contains many photons, the transmitted intensity T_{ab} is proportional to the probability of one “representative” photon incoming along \hat{s}_a , to emerge along \hat{s}_b .

The average transmission coefficient $\overline{T_{ab}}$ is obtained by summing all the possible scattering amplitudes, $A_n^{\{R,m\}}$, corresponding to a given configuration $\{R, m\}$. Here $\{R\}$ accounts for the spatial positions of all scatterers, and $\{m\}$ is a notation for their internal Zeeman states both before and after scattering. The index n denotes one possible multiple scattering path. Squaring the sum of amplitudes we have [8]

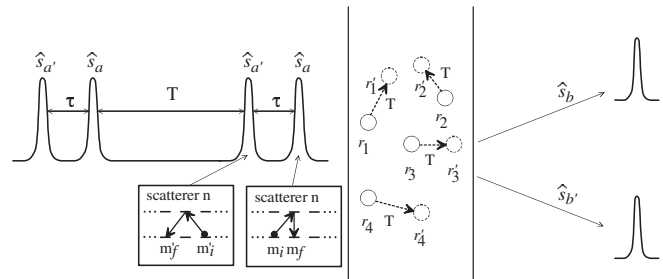


FIG. 1. Photons in each pair of pulses are scattered by atoms at identical positions \mathbf{r}_i but with distinct and uncorrelated quantum numbers (m_i, m_f) and (m_i', m_f') . After a time $T \gg \tau$, a new measurement is performed after the atoms moved to new positions \mathbf{r}_i' .

$$\bar{T}_{ab} = \overline{|\sum_n A_n^{\{R,m\}}|^2} = \sum_{nn'} \overline{A_n^{\{R,m\}} A_{n'}^{\{R,m\}*}} \quad (2)$$

where the overbar denotes a configuration average, over both $\{R\}$ and $\{m\}$. When averaging over $\{R\}$, all cross terms $n \neq n'$ vanish because of large fluctuating phase shifts, so that $\bar{T}_{ab} = \sum_n \overline{|A_n^{\{m\}}|^2}$. This expression, known as the intensity diffuson, is the leading approximation in the weak disorder limit $k_0 l \gg 1$, where k_0 and l are, respectively, the wave number and the elastic mean free path of photons. The (ab) pulse contains many photons, and each of them may change the internal state of atoms. Therefore, if $\{m\}$ (respectively $\{m'\}$) is the atomic internal configuration seen by a representative photon of the (ab) [respectively $(a'b')$] pulse, then we can assume that there is no correlation between $\{m\}$ and $\{m'\}$.

Similarly to the average intensity (2), the correlation of the transmission coefficient is

$$\overline{T_{ab} T_{a'b'}} = \overline{T_{ab}^{\{R,m\}} T_{a'b'}^{\{R,m'\}}} = \sum_{ijkl} \overline{A_i^{\{R,m\}} A_j^{\{R,m\}*} A_k^{\{R,m'\}} A_l^{\{R,m'\}*}} \quad (3)$$

since, as before, the averaging over $\{R\}$ leaves only pairs of amplitudes having exactly opposite phase shifts. To leading order in the weak disorder limit, the only nonvanishing contributions involve two diffusons, i.e., two possible pairings of amplitudes, either $i = j, k = l$, which gives $\bar{T}_{ab} \bar{T}_{a'b'}$, or $i = l, j = k$ so that

$$\overline{\delta T_{ab} \delta T_{a'b'}} = \sum_{ij} \overline{A_i^{\{m\}} A_i^{\{m'\}*} A_j^{\{m'\}} A_j^{\{m\}*}}. \quad (4)$$

The correlation function thus appears as products of two amplitudes, that correspond to different internal configurations $\{m\}$ and $\{m'\}$, but to the same scattering path i (or j). Most of multiple scattering paths i and j do not share common scatterers so that we can average $A_i^{\{m\}} A_i^{\{m'\}*}$ and $A_j^{\{m'\}} A_j^{\{m\}*}$ separately, since these averages are taken upon different atoms, and finally,

$$\overline{\delta T_{ab} \delta T_{a'b'}} = \left| \sum_i \overline{A_i^{\{m\}} A_i^{\{m'\}*}} \right|^2 \quad (5)$$

In the theory of multiple scattering it is helpful to use a continuous description [2]. In this framework, one defines two diffuson functions $\mathcal{D}^{(i,c)}$ by [8]

$$\bar{T}_{ab} = \int d\mathbf{r} d\mathbf{r}' \mathcal{D}^{(i)}(\mathbf{r}, \mathbf{r}') \quad (6)$$

and

$$\overline{\delta T_{ab} \delta T_{a'b'}} = \left| \int d\mathbf{r} d\mathbf{r}' e^{ik_0[\Delta \hat{s}_{a,\mathbf{r}} - \Delta \hat{s}_{b,\mathbf{r}'}]} \mathcal{D}^{(c)}(\mathbf{r}, \mathbf{r}') \right|^2 \quad (7)$$

where $\Delta \hat{s}_{a,b} = \hat{s}_{a,b} - \hat{s}_{a',b'}$. The intensity diffuson $\mathcal{D}^{(i)}(\mathbf{r}, \mathbf{r}')$ is the sum of the terms $|A_n^{\{m\}}(\mathbf{r}, \mathbf{r}')|^2$ between

end points \mathbf{r} and \mathbf{r}' . On the other hand, the correlation diffuson $\mathcal{D}^{(c)}(\mathbf{r}, \mathbf{r}')$ is the sum of the terms $A_i^{\{m\}} A_i^{\{m'\}*}$, i.e., that involve uncorrelated configurations $\{m\}$ and $\{m'\}$.

The two functions $\mathcal{D}^{(i,c)}$ are obtained from the iteration of a proper elementary vertex $\mathcal{V}^{(i,c)}$, that describes the microscopic details of the scattering process. The iteration of the elementary vertex is written symbolically [either for $\mathcal{D}^{(i,c)}$, $\mathcal{V}^{(i,c)}$ we shall denote by \mathcal{D} , \mathcal{V}] as

$$\mathcal{D} = \mathcal{V} + \mathcal{V} \mathcal{W} \mathcal{V} + \dots = \mathcal{V} + \mathcal{D} \mathcal{W} \mathcal{V}. \quad (8)$$

The term \mathcal{V} accounts for a single scattering and $\mathcal{D} \mathcal{W} \mathcal{V}$ represents its iteration. The quantity \mathcal{W} describes the propagation of the photon intensity between successive scattering events and it will be described later on.

The elementary vertex is obtained by the pairing of two scattering amplitudes of a photon by an atom. It is given by [9]

$$\mathcal{V} = \frac{4\pi/l}{2j_g + 1} \sum_{m_i} \langle j_g m_2 | V(\hat{\mathbf{e}}_1, \hat{\mathbf{e}}_2) | j_g m_1 \rangle \times \langle j_g m_4 | V(\hat{\mathbf{e}}_3, \hat{\mathbf{e}}_4) | j_g m_3 \rangle^*, \quad (9)$$

where the operator $V(\hat{\mathbf{e}}', \hat{\mathbf{e}}) = \sum_{m_e} \hat{\mathbf{e}}' \cdot \mathbf{d} | j_e m_e \rangle \times \langle j_e m_e | \mathbf{d} \cdot \hat{\mathbf{e}}$ results from the dipolar interaction energy $-\mathbf{d} \cdot \mathbf{E}$ between atoms and photons, \mathbf{d} being the atomic dipole operator and \mathbf{E} the electric field operator. In the case of $\mathcal{V}^{(c)}$, each one of the two coupled scattering amplitudes in (9) might belong to a distinct atomic configuration, meaning that we must consider two distinct couples of initial ($|j_g m_1\rangle, |j_g m_3\rangle$) and final ($|j_g m_2\rangle, |j_g m_4\rangle$) atomic states, as well as two initial ($\hat{\mathbf{e}}_1, \hat{\mathbf{e}}_3$) and final ($\hat{\mathbf{e}}_2, \hat{\mathbf{e}}_4$) photon polarization states. The summations over the quantum numbers m_i result from averaging over initial atomic states and from nondetected final states. Thus $\mathcal{V}^{(c)}$ corresponds to the most general case regarding the m_i quantum numbers. In contrast, $\mathcal{V}^{(i)}$ corresponds to the differential scattering cross section, for which we set $m_1 = m_3, m_2 = m_4, \hat{\mathbf{e}}_1 = \hat{\mathbf{e}}_3$, and $\hat{\mathbf{e}}_2 = \hat{\mathbf{e}}_4$ in (9). This is because the intensity diffuson is built out of two coupled amplitudes that must belong to the same scattering process. This distinction between $\mathcal{V}^{(i)}$ and $\mathcal{V}^{(c)}$, and therefore between $\mathcal{D}^{(i)}$ and $\mathcal{D}^{(c)}$, occurs only for $j_g > 0$ and it is at the basis of the new results we obtain here for mesoscopic speckle correlations.

The iteration (8) is implemented using the decomposition of the various terms into standard basis components, thus leading to the definition of rank four tensors such as

$$\mathcal{V} = \sum_{\alpha\beta\gamma\delta} (\hat{\mathbf{e}}_1)_{-\alpha} (\hat{\mathbf{e}}_2)_{\gamma}^* (\hat{\mathbf{e}}_3)_{-\beta}^* (\hat{\mathbf{e}}_4)_{\delta} \mathcal{V}_{\alpha\beta\gamma\delta}. \quad (10)$$

Likewise, the iteration Eq. (8) for the diffusons acquires a tensorial structure, which reads

$$\mathcal{D}_{\alpha\beta,\gamma\delta} = \mathcal{V}_{\alpha\beta,\gamma\delta} + W \sum_{\mu\nu\rho\sigma} \mathcal{D}_{\alpha\beta,\mu\nu} b_{\mu\nu,\rho\sigma} \mathcal{V}_{\rho\sigma,\gamma\delta}. \quad (11)$$

Here $\mathcal{W} = Wb$, the function W describes the scalar part of the photon intensity propagator and b , defined by

$$b_{\alpha\beta,\gamma\delta} = \langle [\delta_{\alpha\gamma} - (-1)^\gamma \hat{s}_\alpha \hat{s}_{-\gamma}] [\delta_{\beta\delta} - (-1)^\beta \hat{s}_{-\beta} \hat{s}_\delta] \rangle, \quad (12)$$

accounts for the polarization dependent part. This expression follows at once by noticing that after being scattered by an atom, the two outgoing amplitudes propagate with a wave vector $\hat{\mathbf{s}} = \mathbf{k}/k_0$, random in direction but identical for both, and with two different polarization components. Since $\hat{\mathbf{s}}$ is random, the intensity propagation is averaged $\langle \dots \rangle$ over photon wave vectors direction. The term $\delta_{\mu\nu} - (-1)^\nu \hat{s}_\mu \hat{s}_{-\nu}$ expresses transversality.

To proceed further, we use the Wigner-Eckart theorem to rewrite the tensor $\mathcal{V}_{\alpha\beta,\gamma\delta}$ in terms of a summation of product of $3j$ symbols,

$$\begin{aligned} \mathcal{V}_{\alpha\beta,\gamma\delta} &= 3(2j_e + 1) a_{j_g j_e} \sum_{m_i m_e m'_e} \begin{pmatrix} j_e & 1 & j_g \\ -m_e & \alpha & m_1 \end{pmatrix} \\ &\times \begin{pmatrix} j_e & 1 & j_g \\ -m_e & \gamma & m_2 \end{pmatrix} \begin{pmatrix} j_e & 1 & j_g \\ -m'_e & \delta & m_4 \end{pmatrix} \\ &\times \begin{pmatrix} j_e & 1 & j_g \\ -m'_e & \beta & m_3 \end{pmatrix}, \end{aligned} \quad (13)$$

where $a_{j_g j_e} = (2j_e + 1)/3(2j_g + 1)$. The two tensors $b_{\alpha\beta,\gamma\delta}$ and $\mathcal{V}_{\alpha\beta,\gamma\delta}$ can be written in the form of a 9×9 matrix. According to the spectral decomposition theorem, they can be decomposed using an orthonormal set of (generally) nine projectors $T^{(K)}$ [10]. Looking at (11), we wish to find the spectral decomposition of \mathcal{D} using the spectral decomposition of \mathcal{V} and $b\mathcal{V}$. The problem is that they do not share the same projectors set in their spectral decomposition. We are thus led to define a new tensor U by $\mathcal{D} = U\mathcal{V}$. It obeys the iteration equation $U = 1 + WU\mathcal{V}b$ and it involves only the spectral decomposition of $\mathcal{V}^{(i,c)}b = \sum_{K=0}^8 u_K^{(i,c)} T^{(K)}$. This leads immediately to

$$\mathcal{D}_{\alpha\beta,\gamma\delta}^{(i,c)} = \sum_K U_K^{(i,c)} (\mathcal{V}_K^{(i,c)})_{\alpha\beta,\gamma\delta} \quad (14)$$

with $\mathcal{V}_K^{(i,c)} = T^{(K)} \mathcal{V}^{(i,c)}$ and

$$U_K^{(i,c)} = \frac{4\pi/l}{1 - W(q)u_K^{(i,c)}} \simeq \frac{8\pi c}{3l^2} a_{j_g j_e} \frac{1/u_K^{(i,c)}}{\frac{1}{\tau_K^{(i,c)}} + Dq^2}, \quad (15)$$

where \mathbf{q} (with $q = |\mathbf{q}|$) is the Fourier variable of the difference $\mathbf{R} = \mathbf{r}' - \mathbf{r}$ between the two end points of a multiple scattering sequence. The right-hand side in (15) is obtained by using the diffusion approximation (i.e., $ql \ll 1$), so that $W(q) \simeq \frac{3}{2a_{j_g j_e}} (1 - q^2 l^2/3)$, where $D = cl/3$ is the photon diffusion coefficient [2] and c the speed of light.

We identify the set of characteristic times

$$\tau_K^{(i,c)} = \left(\frac{l}{c}\right) \frac{u_K^{(i,c)}}{\frac{2}{3} a_{j_g j_e} - u_K^{(i,c)}}. \quad (16)$$

For $\mathcal{V}^{(i)}$ [$m_1 = m_3, m_2 = m_4$ in (13)], it is straightforward to check that $(\mathcal{V}^{(i)}b)_{\alpha\beta,\gamma\delta}$ admits a spectral decomposition over 3 projectors $T^{(K)}$ only [2,6]. In contrast, for $\mathcal{V}^{(c)}$ there are no constraint on the m_i quantum numbers, so that the total angular momentum needs not to be conserved, and the corresponding spectral decomposition involves usually more than 3 projectors $T^{(K)}$. For a non-degenerate ground state level ($j_g = 0$), angular momentum is automatically conserved and the two vertices become identical, $\mathcal{V}^{(c)} = \mathcal{V}^{(i)}$.

The poles that occur in (15) correspond to diffusive modes of lifetime $\tau_K^{(i,c)}$. This shows up when rewriting (14), with the help of (10), in real space

$$\mathcal{D}^{(i,c)}(\mathbf{r}, \mathbf{r}') = \sum_K \frac{Y_K^{(i,c)}}{u_K^{(i,c)}} \int_0^\infty dt \mathcal{D}(\mathbf{r}, \mathbf{r}', t) e^{-t/\tau_K^{(i,c)}}, \quad (17)$$

where

$$Y_K^{(c)} = \sum_{\alpha\beta\gamma\delta} (\hat{\mathbf{e}}_a)_{-\alpha} (\hat{\mathbf{e}}_b)_\gamma (\hat{\mathbf{e}}_{a'})_{-\beta} (\hat{\mathbf{e}}_{b'})_\delta (\mathcal{V}_K^{(c)})_{\alpha\beta,\gamma\delta} \quad (18)$$

and a corresponding expression for $Y_K^{(i)}$ obtained by setting $a' = a, b' = b$ and $\mathcal{V}^{(i)}$ in the previous relation. The scalar diffusion propagator $\mathcal{D}(\mathbf{r}, \mathbf{r}', t)$ obeys a diffusion equation whose solution for a slab of width L is well known [2,10] and leads for (6) and (7) to

$$\bar{T}_{ab} = \sum_{K=0}^2 \frac{Y_K^{(i)}}{u_K^{(i)}} \mathcal{D}(Q_K^{(i)}(0)) \quad (19)$$

and

$$\overline{\delta T_{ab} \delta T_{a'b'}} = \delta_{\Delta\hat{s}_a, \Delta\hat{s}_b} \left[\sum_{K=0}^8 \frac{Y_K^{(c)}}{u_K^{(c)}} \mathcal{D}(Q_K^{(c)}(q_p)) \right]^2. \quad (20)$$

We have defined the quantities $q_p = k_0 \Delta\hat{s}_a$, $Q_K^{(i,c)}(x) = \sqrt{x^2 + (1/D\tau_K^{(i,c)})}$, and $\mathcal{D}(x) = \sinh^2(xl)/[xl \sinh(xL)]$.

We now analyze expressions (19) and (20), which constitute the main results of this Letter. First, consider the modes of the average intensity $\mathcal{D}^{(i)}$. It is easy to check that $\tau_0^{(i)}$ is infinite as a result of the Ward identity $u_0^{(i)} = \frac{2}{3} a_{j_g j_e}$ [see (16)]. The corresponding Goldstone mode $U_0^{(i)} \propto 1/Dq^2$ expresses energy conservation and long-range propagation of the average intensity. The two other modes $U_K^{(i)}$ are exponentially damped with the times $\tau_K^{(i)}$ [see (17)]. This expresses photon depolarization in multiple scattering [2,6,7,11].

The spectral decomposition of $\mathcal{D}^{(c)}$ gives rise to nine modes and their corresponding times $\tau_K^{(c)}$. Such times are

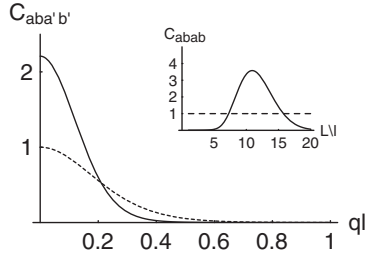


FIG. 2. Angular correlation function $C_{aba'b'}$ plotted as a function of $q = k_0 \Delta \hat{s}_a$ for an atomic transition between nondegenerate ($j_g = 0, j_e = 1$, dashed line) and degenerate energy levels ($j_g = 1, j_e = 2$, solid line). The nondegenerate case describes the classical Rayleigh scattering of a polarized wave. These curves correspond to $L = 7l$. The inset gives the dependence of intensity fluctuations C_{abab} upon the width L (the dashed line is the Rayleigh law).

well known to occur in quantum mesoscopic physics, e.g., in conductance fluctuations of disordered metals in the presence of magnetic impurities [2,12]. The times $\tau_K^{(c)}$ describe how underlying interferences between multiply scattered waves (electrons in metals, photons in the present case) are washed out in the presence of other degrees of freedom. The surprising and new feature of the atom-photon scattering is the occurrence of a mode ($K = 0$) with a negative $\tau_0^{(c)}$. According to (17), this corresponds to an amplified mode that enhances the angular correlation function. This amplified mode is present for a degenerate atomic transition ($j_g, j_e > 0$) and vanishes otherwise. Its origin can be traced out from the vertex (9) which can be written as a sum of an incoherent contribution (i.e., a sum of probabilities) present both in $\mathcal{V}^{(i)}$ and $\mathcal{V}^{(c)}$, and a coherent contribution (i.e., a sum of products of quantum amplitudes associated to different Zeeman states). The coherent contribution enhances $\mathcal{V}^{(c)}$ with respect to $\mathcal{V}^{(i)}$ and its iteration gives rise to the amplified mode characterized by $\tau_0^{(c)}$. Expressions (19) and (20) lead to an expression of $C_{aba'b'}$ plotted in Fig. 2. This expression allows us to recover the limiting case of a scalar classical wave [1] corresponding to $Y^{(c)} = Y^{(i)} = 1$ and to a single mode with infinite $\tau_0^{(i,c)}$. It also provides a simple expression for angular speckle correlations of classical Rayleigh scattering of a polarized wave [10,13]. For atomic transitions between degenerate levels, $C_{aba'b'}$ exhibits a steep decrease and large intensity fluctuations (see Fig. 2) as compared to the case of nondegenerate atomic levels. Intensity fluctuations measured by C_{abab} become larger than 1, unlike Rayleigh law, $C_{abab} = 1$, well obeyed by classical scatterers. Large intensity fluctuations result from the amplified mode $\tau_0^{(c)}$ which leads to a divergence of the integral in (17). This divergence is cutoff by other dephasing mechanisms, such as Doppler shift, inelastic scattering or finite

size of the atomic trap. Denoting by Λ this upper cutoff, and assuming that the dominant contributions to \overline{T}_{ab} and to $\overline{\delta T_{ab}^2}$ are given, respectively, by the Goldstone and the amplified modes, we deduce from (17) that

$$C_{abab} = \frac{\overline{\delta T_{ab}^2}}{\overline{T}_{ab}^2} \simeq A \pi^4 \left(\frac{e^{X\Lambda D/L^2} - 1}{X} \right)^2, \quad (21)$$

where $X \equiv (L/L_0^{(c)})^2 - \pi^2$ involves the diffusion length $L_0^{(c)} \equiv \sqrt{D|\tau_0^{(c)}|}$ and A is a constant equal to one for a nondegenerate ground state. This approximate expression reproduces the main features of C_{abab} plotted in the inset of Fig. 2, namely, that it can be larger than the Rayleigh term and that it is peaked at a value of L that depends on the cutoff Λ . When $L_0^{(c)}$ and Λ are infinite, C_{abab} becomes independent of L and is given by the Rayleigh law. Relative fluctuations as given by (21) thus provide a direct probe of dephasing mechanisms in cold atomic gases.

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