Supplemental Material: Coherent scattering of near-resonant light by a Dense Microscopic Cold Atomic cloud

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This Supplemental Material presents more details about the definition of the coherent transfer function $\mathcal{S}(\omega)$ and the expressions of the laser field used in the modeling. It also derives the mean-field expression of the susceptibility for a multi-level alkali atom that we use in the Lorentz-Lorenz formula to calculate the field scattered by the cloud. Finally, we discuss the influence on the line shape of a misalignment of the laser probe with respect to the atomic cloud.

I. COHERENT TRANSFER FUNCTION

The experimental configuration used in the experiment gives access to the overlap between the total field in the forward direction $\mathbf{E} = \mathbf{E}_{\rm L} + \mathbf{E}_{\rm sc}$ and the mode $\mathbf{g}$ of the single-mode-fibered detector

$$\mathcal{E}(\omega) = \int \mathbf{E}(\mathbf{r},\omega)\cdot\mathbf{g}^*(\mathbf{r})dS \tag{1}$$

with dS a differential area element perpendicular to the optical axis. As the fiber mode is matched to the incoming light, $\mathbf{g} \propto \mathbf{E}_{\rm L}$, and the total transfer function is

$$\mathcal{S}_{\rm tot}(\omega) = \frac{\mathcal{E}(\omega)}{\mathcal{E}_{\rm L}(\omega)} = \frac{\int \mathbf{E}(\mathbf{r},\omega)\cdot\mathbf{E}_{\rm L}^*(\mathbf{r})dS}{\int|\mathbf{E}_{\rm L}(\mathbf{r})|^2 dS}\ . \tag{2}$$

We then decompose the scattered field into the coherent and incoherent (fluctuating) components: $\mathbf{E}_{\rm sc} = \langle\mathbf{E}_{\rm sc}\rangle + \delta\mathbf{E}_{\rm sc}$, where $\langle\cdot\rangle$ indicates an average over many spatial configurations of the cloud. In the experiment, we measure the configuration-averaged quantity

$$\begin{aligned}\langle|\mathcal{E}(\omega)|^2\rangle &\propto \left|\int(\mathbf{E}_{\rm L}+\langle\mathbf{E}_{\rm sc}\rangle)\cdot\mathbf{E}_{\rm L}^* dS\right|^2 \\ &+ \langle\left|\int\delta\mathbf{E}_{\rm sc}\cdot\mathbf{E}_{\rm L}^* dS\right|^2\rangle\ ,\end{aligned} \tag{3}$$

(taking into account $\langle\delta\mathbf{E}_{\rm sc}\rangle = 0$), which we cast in the form $|\mathcal{E}_{\rm coh}(\omega)|^2 + \langle|\mathcal{E}_{\rm incoh}(\omega)|^2\rangle$. As explained in the main text, $|\mathcal{E}_{\rm coh}(\omega)|^2 \gg \langle|\mathcal{E}_{\rm incoh}(\omega)|^2\rangle$ in the direction of propagation of the laser, and therefore, we measure essentially the coherent part to the total transfer function. We thus define the coherent optical transfer function:

$$\mathcal{S}(\omega) = \frac{\langle\mathcal{E}(\omega)\rangle}{\mathcal{E}_{\rm L}(\omega)} = 1 + \frac{\int\langle\mathbf{E}_{\rm sc}\rangle\cdot\mathbf{E}_{\rm L}^* dS}{\int|\mathbf{E}_{\rm L}|^2 dS}\ . \tag{4}$$

In general, the transfer function defined above does not correspond to the transmission of the cloud defined as

$$T(\omega) = \frac{\langle\int|\mathbf{E}(\mathbf{r},\omega)|^2 dS\rangle}{\int|\mathbf{E}_{\rm L}(\mathbf{r})|^2 dS}\ . \tag{5}$$

If the solid angle of the collecting lens L1 is very small, $T(\omega)$ and $|\mathcal{S}_{\rm tot}(\omega)|^2$ coincide. Otherwise, the relation between the transfer function and the transmission can be found by decomposing, once again, the total field as $\mathbf{E} = \mathbf{E}_{\rm L} + \langle\mathbf{E}_{\rm sc}\rangle + \delta\mathbf{E}_{\rm sc}$. We then get

$$\begin{aligned}T(\omega) &= |\mathcal{S}(\omega)|^2 + \frac{\int\langle|\delta\mathbf{E}_{\rm sc}|^2\rangle dS}{\int|\mathbf{E}_{\rm L}(\mathbf{r})|^2 dS} \\ &+ \frac{\int|\langle\mathbf{E}_{\rm sc}\rangle|^2 dS}{\int|\mathbf{E}_{\rm L}(\mathbf{r})|^2 dS} - \left|\frac{\int\langle\mathbf{E}_{\rm sc}\rangle\cdot\mathbf{E}_{\rm L}^* dS}{\int|\mathbf{E}_{\rm L}(\mathbf{r})|^2 dS}\right|^2\ .\end{aligned} \tag{6}$$

Using the Cauchy-Schwartz inequality

$$\left|\int\langle\mathbf{E}_{\rm sc}\rangle\cdot\mathbf{E}_{\rm L}^* dS\right|^2 \le \int|\langle\mathbf{E}_{\rm sc}\rangle|^2 dS\int|\mathbf{E}_{\rm L}(\mathbf{r})|^2 dS$$

yields $|\mathcal{S}(\omega)|^2 \le T(\omega)$.

II. DESCRIPTION OF THE PROBE LASER FIELD

In the theoretical models (Lorentz local-field and microscopic), we use the following paraxial approximation for the amplitude of the x component of the electric probe laser field (with a waist w, see Fig. 1a in main text):

$$E_{{\rm L},x}(x,y,z) = \frac{E_0}{1+i\frac{z}{z_{\rm R}}}\exp\left[ik\frac{x^2+y^2}{2q(z)}\right]\exp[ikz]\ , \tag{7}$$

with $\frac{1}{q(z)} = \frac{1}{R(z)} + \frac{2\,i}{kw^2(z)}$, $z_{\rm R} = kw^2/2$ the Rayleigh length, $w(z) = w\sqrt{1+z^2/z_{\rm R}^2}$ and $R(z) = z + z_{\rm R}^2/z$.

As is well-known, this paraxial expression is not an exact solution of Maxwell's equation. We have therefore checked, for the calculation of the transfer function based on the Lorentz-Lorenz formula, that the field scattered by the cloud using the paraxial approximation is numerically very close to the one obtained by using the plane wave decomposition of the incident beam (which corresponds to an exact solution of Maxwell's equations).

III. DERIVATION OF THE MEAN-FIELD SUSCEPTIBILITY FOR A MULTI-LEVEL ATOM

In this section we derive the mean-field electric susceptibility for a multi-level atom, when the recurrent scattering contributions are ignored. This is then used in the main section of the paper to calculate the mean-field response and the "cooperative Lamb shift" for a ^{87}Rb $F = 2$ ground-state manifold that differ from the ones obtained for an isotropic $F = 0 \to F' = 1$ transition.

We use the general formalism of Ref. [1] and derive the multi-level electric susceptibility as in Ref. [2]. We consider an atom with ground states $|g,\nu\rangle$ and excited states $|e,\eta\rangle$. Here ν and η represent the Zeeman sub-levels of the ground and excited states, respectively, separated by a transition at a frequency ω_0.

The positive frequency component of the electric field amplitude is the sum of the incident coherent field $\mathbf{D}_F^+(\mathbf{r})$ and the scattered field from the atomic polarization $\hat{\mathbf{P}}^+(\mathbf{r})$,

$$\epsilon_0\hat{\mathbf{E}}^+(\mathbf{r}) = \mathbf{D}_F^+(\mathbf{r}) + \int d^3r'\,\mathsf{G}(\mathbf{r}-\mathbf{r}')\,\hat{\mathbf{P}}^+(\mathbf{r}')\,. \quad (8)$$

The monochromatic dipole radiation kernel $\mathsf{G}(\mathbf{r})$ [3] gives the radiated field at $\mathbf{r}$ from a dipole with the amplitude $\hat{\mathbf{d}}$ residing at the origin:

$$\mathsf{G}(\mathbf{r})\,\hat{\mathbf{d}} = \frac{k^3}{4\pi}\Big\{(\hat{\mathbf{n}}\times\hat{\mathbf{d}})\times\hat{\mathbf{n}}\frac{e^{ikr}}{kr} + [3\hat{\mathbf{n}}(\hat{\mathbf{n}}\cdot\hat{\mathbf{d}})-\hat{\mathbf{d}}]\big[\frac{1}{(kr)^3}-\frac{i}{(kr)^2}\big]e^{ikr}\Big\} - \frac{\hat{\mathbf{d}}\,\delta(\mathbf{r})}{3}\,, \quad (9)$$

where $k = \omega/c$, ω denotes the laser light frequency, and $\hat{\mathbf{n}} = \mathbf{r}/r$.

Using the second quantized atomic field operators $\hat\psi_{g\nu}(\mathbf{r})$ and $\hat\psi_{e\eta}(\mathbf{r})$, the positive frequency component of the atomic polarization density can be written in terms of contributions from different sub-level transitions as

$$\hat{\mathbf{P}}^+(\mathbf{r}) = \sum_{\nu,\eta}\hat{\mathbf{P}}^+_{\nu\eta}(\mathbf{r})\,, \quad (10)$$

$$\hat{\mathbf{P}}^+_{\nu\eta}(\mathbf{r}) \equiv \mathbf{d}_{g\nu e\eta}\hat\psi^\dagger_{g\nu}(\mathbf{r})\hat\psi_{e\eta}(\mathbf{r})\,, \quad (11)$$

where $\mathbf{d}_{g\nu e\eta}$ represents the dipole matrix element for the transition $|e,\eta\rangle \to |g,\nu\rangle$

$$\mathbf{d}_{g\nu e\eta} \equiv \mathcal{D}\sum_\sigma \hat{\mathbf{e}}_\sigma\langle e\eta;1g|1\sigma;g\nu\rangle \equiv \mathcal{D}\sum_\sigma\hat{\mathbf{e}}_\sigma\mathcal{C}^{(\sigma)}_{\nu,\eta}\,. \quad (12)$$

Here the sum is over the unit circular polarization vectors $\sigma = \pm1, 0$, and $\mathcal{C}^{(\sigma)}_{\nu,\eta}$ denote the Clebsch-Gordan coefficients of the corresponding optical transitions. The reduced dipole matrix element $\mathcal{D}$ is related to the linewidth of the transition Γ by

$$\Gamma = \frac{\mathcal{D}^2\omega_0^3}{3\pi\hbar\epsilon_0 c^3}\,, \quad (13)$$

and $\mathbf{d}_{e\eta g\nu} = \mathbf{d}^*_{g\nu e\eta}$. The light fields with the polarizations $\hat{\mathbf{e}}_\pm$ and $\hat{\mathbf{e}}_0$ drive the transitions $|g,\nu\rangle \to |e,\nu\pm1\rangle$ and $|g,\nu\rangle \to |e,\nu\rangle$, respectively, in such a way that only the terms $\sigma = \eta-\nu$ in Eq. (12) are nonvanishing.

As described in Refs. [1, 2], in the limit of low light intensity we obtain the equation of motion for the expectation value of the polarization component $\mathbf{P}^+_{\nu\eta}(\mathbf{r}) = \langle\hat{\mathbf{P}}^+_{\nu\eta}(\mathbf{r})\rangle$,

$$\frac{d}{dt}\mathbf{P}^+_{\nu\eta}(\mathbf{r}) = (i\bar\Delta_{g\nu e\eta}-\frac{\Gamma}{2})\mathbf{P}^+_{\nu\eta}(\mathbf{r}) + i\xi\rho_\nu(\mathbf{r})\mathsf{P}^{\nu\eta}_{\eta\nu}\mathbf{D}_F^+(\mathbf{r}) + i\xi\int d^3r'\,\mathsf{P}^{\nu\eta}_{\eta\tau}\mathsf{G}(\mathbf{r}-\mathbf{r}')\,\langle\hat\psi^\dagger_{g\nu}(\mathbf{r})\hat{\mathbf{P}}^+(\mathbf{r}')\hat\psi_{g\tau}(\mathbf{r})\rangle\,, \quad (14)$$

where we assumed that there are no ground-state coherences between the different hyperfine states, so that $\langle\hat\psi^\dagger_{g\nu}\hat\psi_{g\tau}\rangle = \delta_{\nu,\tau}\rho_\nu$, with ρ_ν the atom denisty of the ground state $|g,\nu\rangle$.

We have defined $\xi = \mathcal{D}^2/(\hbar\epsilon_0)$ and $\Delta_{a\nu b\eta} = \Delta_{b\eta}-\Delta_{a\nu}$ $(a,b = g,e)$. In the presence of a magnetic field B, the atom-light detuning is $\Delta_{e\eta} = \omega-(\omega_0+\mu_B B g_l^{(e)}\eta/\hbar)$ and $\Delta_{g\nu} = -\mu_B B g_l^{(g)}\nu/\hbar$, with ω denoting the frequency of the incident light, and $g_l^{(g)}$ and $g_l^{(e)}$ the Landé factors of the ground and excited states, respectively. In Eq. (14) we also introduced the tensor

$$\mathsf{P}^{\nu\eta}_{\mu\tau} \equiv \frac{\mathbf{d}_{g\nu e\eta}\mathbf{d}_{e\mu g\tau}}{\mathcal{D}^2} = \sum_{\sigma,\varsigma}\hat{\mathbf{e}}_\sigma\hat{\mathbf{e}}^*_\varsigma\mathcal{C}^{(\sigma)}_{\nu,\eta}\mathcal{C}^{(\varsigma)}_{\tau,\mu}\,. \quad (15)$$

The pair correlation function $\langle\hat\psi^\dagger_{g\nu}(\mathbf{r})\hat{\mathbf{P}}^+(\mathbf{r}')\hat\psi_{g\tau}(\mathbf{r})\rangle$ describes light-induced correlations between the atoms at $\mathbf{r}$ and $\mathbf{r}'$ [1]. These correlations are non-trivial in the presence of recurrent scattering processes. In the mean-field theory we neglect recurrent scattering by the decorrelation approximation $\langle\hat\psi^\dagger_{g\nu}(\mathbf{r})\hat{\mathbf{P}}^+(\mathbf{r}')\hat\psi_{g\tau}(\mathbf{r})\rangle \simeq \rho_\nu(\mathbf{r})\mathbf{P}^+(\mathbf{r}')$ in Eq. (14).

We are interested in the steady-state solution of the resulting approximation to Eq. (14). In the interaction potential between the two atoms in Eq. (14), we can now remove the contact term $\mathsf{G}(\mathbf{r}) \to \mathsf{G}(\mathbf{r})+\delta(\mathbf{r})/3$, and then eliminate the scattered field between Eqs. (14) and (8) (this procedure can be derived rigorously [4]). We obtain

$$\mathbf{P}_{\nu\eta} = \alpha_{\nu\eta}\rho_\nu\mathsf{P}^{\nu\eta}_{\eta\nu}(\epsilon_0\mathbf{E}+\mathbf{P}/3)\,, \quad (16)$$

where the polarizability is given by

$$\alpha_{\nu\eta} = -\frac{\mathcal{D}^2}{\hbar\epsilon_0}\frac{1}{\Delta_{g\nu e\eta}+i\frac{\Gamma}{2}}\,. \quad (17)$$

Equation (16) now leads to a coupled set of linear equations between the different polarization components [2] that can be solved

$$\mathbf{P}_{\nu\eta} = \frac{\alpha_{\nu\eta}\rho_\nu[\mathcal{C}^{(\sigma')}_{\nu,\eta}]^2}{1-\sum_{\tau,\zeta}\alpha_{\tau\zeta}\rho_\tau[\mathcal{C}^{(\sigma')}_{\tau,\zeta}]^2/3}\,\epsilon_0(\hat{\mathbf{e}}^*_{\sigma'}\cdot\mathbf{E})\hat{\mathbf{e}}_{\sigma'}\,, \quad (18)$$

where $\sigma' = \eta - \nu$. The sum in the denominator therefore includes the components for which $\mathcal{C}^{(\sigma')}_{\tau,\zeta} \neq 0$, i.e., the components for which $\mathbf{P}_{\tau\zeta}$ is parallel to $\mathbf{P}_{\nu\eta}$.

We can then separate the different vector components of the total polarization $\mathbf{P} = \sum_{\nu,\eta} \mathbf{P}_{\nu\eta}$ by considering each value of the spherical polarization component σ' separately

$$\hat{\mathbf{e}}^*_{\sigma'} \cdot \mathbf{P} = \frac{\sum_{\nu,\eta} \alpha_{\nu\eta}\rho_\nu [\mathcal{C}^{(\sigma')}_{\nu,\eta}]^2}{1 - \sum_{\tau,\zeta} \alpha_{\tau\zeta}\rho_\tau [\mathcal{C}^{(\sigma')}_{\tau,\zeta}]^2/3}\, \epsilon_0 (\hat{\mathbf{e}}^*_{\sigma'} \cdot \mathbf{E})\,. \quad (19)$$

This now gives the electric susceptibility for $\sigma' = \pm 1, 0$.

To describe the experiment (for which the magnetic field is $B \approx 0$ G), we consider a linearly polarized laser beam driving π transitions ($\sigma = 0$). We also assume all the Zeeman states of the ground level $F = 2$ equally populated, implying $\rho_\nu = \rho_{\text{total}}/5$. In this way, the only effect of the internal structure of the atom is to multiply the polarizability given by Eq. (17) (with $\Delta_{g\nu e\eta} = \Delta = \omega - \omega_0$) by $\sum_\nu [\mathcal{C}^{(0)}_{\nu,\nu}]^2/5 = 7/15$, as written in the main text.

IV. INFLUENCE OF A MISALIGNMENT OF THE PROBE WITH RESPECT TO THE CLOUD

In this last section, we analyze the influence of a misalignement of the probe with respect to the atomic cloud. The dimensions of the cloud are $(a_\perp, a_z) = (0.2, 1.2)\,\mu$m, to be compared to the probe beam waist $w = 1.20 \pm 0.05\,\mu$m. These very small numbers indicate that the perfect alignment of the probe with respect to the cloud is challenging. As demonstrated in [5], a longitudinal displacement of the probe focal point with respect to the center of the cloud leads to asymmetric atomic line shapes in transmission. Reference [5] interprets this asymmetry as a lensing effect induced by the cloud.

We have modeled the effect of a misalignment of the probe using the coupled dipole equations. We calculate the transfer function as described in the main text for different transverse and longitudinal positions of the cloud with respect to the probe beam. Figure 1a presents the results for a longitudinal displacement of the cloud along the direction of propagation of the probe beam for $N = 20$ atoms. We observe a strong asymmetry of the line shape when the cloud is displaced by $\pm z_{\text{R}}$ with respect to the position of the beam waist. This asymmetry can be understood as an effect of the Gouy phase of the probe laser (see below).

Finally, we displace transversally the atomic cloud in the focal plane of the laser beam, perpendicular to the optical axis. The results are shown in Fig. 1b. The main effect of the displacement is to reduce the amplitude of the line, without inducing any frequency shift or any change in the linewidth. We have tested that this conclusion remains valid for all atom numbers investigated in this work. In particular, the transverse displacement does not induce any extra asymmetry in the line shape with respect to the case of an unshifted position of the cloud.

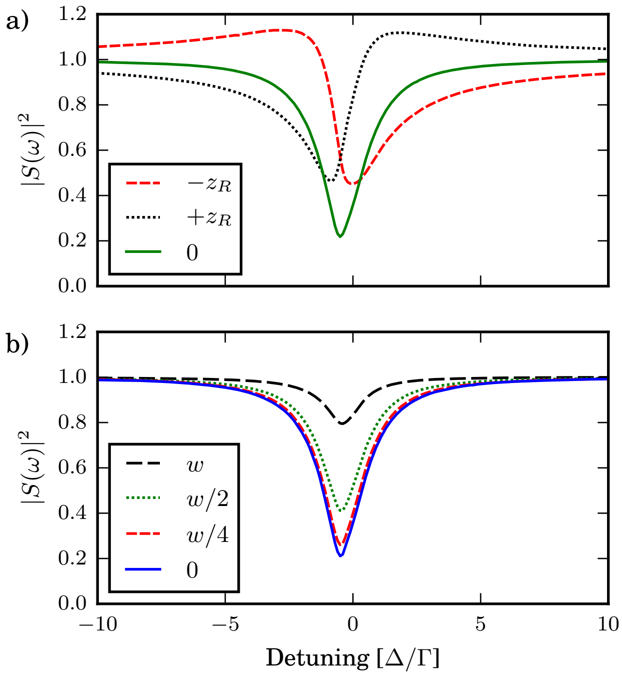

FIG. 1: (a) Influence of a longitudinal displacement of the position of the waist of the probe beam with respect to the center of the atomic cloud. The focus of the beam is displaced by $z_0 = \pm z_{\text{R}}$, with respect to the position of the center of the cloud ($z_0 = 0$). (b) Influence of a transverse misalignment of the probe beam with respect to the center of the atomic cloud. We considered 3 transverse displacements with respect to the center of the cloud (0): $w/4$, $w/2$ and w, where $w = 1.2\,\mu$m is the probe beam waist. Both (a) and (b) correspond to $N = 20$.

Both effects can be understood using the following simplified model. Let us consider the case of a dielectric cloud with a size smaller than $1/k$ (k is the wave-vector of the light) with a polarizability $\alpha(\omega)$ (with a width Γ_{c} and a central frequency ω_{c}), located at a position (x_0, y_0, z_0) around the focal point of the probe beam at position $(0, 0, 0)$. At a distance $z \gg |x_0|, |y_0|, |z_0|$ in the far-field, the component of the electric field along the x-axis scattered by the cloud in the direction of the propagation of the probe (z axis) is [3]:

$$E_{\text{sc},x}(z) \approx \frac{k^2}{4\pi}\alpha(\omega)|E_{\text{L}}(x_0, y_0, z_0)|e^{i[\psi(z_0)+kz_0]} \times \frac{e^{ik\sqrt{(z-z_0)^2+x_0^2+y_0^2}}}{z}\,, \quad (20)$$

with $\psi(z_0)$ the Gouy phase given by $\psi(z_0) = -\arctan[z_0/z_{\text{R}}]$. Using the far-field expression of the laser electric field (7), we get the total field in the direction of propagation of the probe:

$$E_{\mathrm{tot},x} = -i\frac{z_{\mathrm{R}}}{z}|E_{\mathrm{L}}(0)|e^{ikz} \times \left(1 + i\frac{k^2\alpha(\omega)}{4\pi z_{\mathrm{R}}}\frac{|E_{\mathrm{L}}(x_0,y_0,z_0)|}{|E_{\mathrm{L}}(0)|}e^{i\psi(z_0)}\right) . \quad (21)$$

Along the axis, the transfer function is:

$$\frac{E_{\mathrm{tot},x}}{E_{\mathrm{L},x}} = 1 + i\frac{k^2\alpha(\omega)}{4\pi z_{\mathrm{R}}}\frac{|E_{\mathrm{L}}(x_0,y_0,z_0)|}{|E_{\mathrm{L}}(0)|}e^{i\psi(z_0)} , \quad (22)$$

which can be cast in the form of an on-axis transfer function:

$$\mathcal{S}_{\mathrm{axis}}(\omega) = 1 - \frac{A}{1-2i\frac{\omega-\omega_c}{\Gamma_c}}e^{i\psi(z_0)} , \quad (23)$$

with A a real number This expression shows that for a cloud located at $(x_0, y_0, 0)$ (transverse displacement), the amplitude of the extinction is attenuated by the ratio $|E_L(x_0, y_0, z_0)|/|E_L(0)|$, as observed in Fig. 1(b). When the cloud is centered in $(0, 0, z_0)$ (with z_0 small but not negligible with respect to z_{R}), the phase factor $e^{i\psi(z_0)}$ involving the Gouy phase leads to an asymmetric function of ω: this explains the asymmetry observed in Fig. 1a. From the very slight asymmetry observed on the data (see Fig. 1a of the main text), we infer the phase $\psi(z_0)$ by fitting the data by the expression $|\mathcal{S}_{\mathrm{axis}}(\omega)|^2$ (Eq. 23). We find $\psi(z_0) \approx 0.3\,\mathrm{rad}$ and extract a longitudinal displacement of the cloud $z_0 \approx 1.8\,\mu\mathrm{m}$. Importantly, we find that this phase is independent of the atom number, as it should if it is a geometric phase. A lensing effect would lead to an asymmetry that would vary with the number of atoms.

We conclude from this study that a misalignment of the probe cannot introduce any narrowing of the line or any extra shift, nor wash out any double structure. The misalignment thus cannot explain the discrepancy between the theoretical models and the data, which systematically feature a smaller shift and linewidth and no significant asymmetry.

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