Thin film of two-level atoms: a simple model of optical bistability and self-pulsation

A. M. Basharov

Engineering-Physics Institute, Moscow

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Bistable and periodically pulsating reflection of a monochromatic optical wave is predicted for a thin film of resonant atoms deposited on an insulating substrate. The correspondence between these effects and the transmission of a monochromatic wave by an optical resonator is considered.

A thin film of two-level atoms of thickness much less than the wavelength of the incident light represents a very comprehensive model for studies of nonlinear optical phenomena such as nonlinear surface waves,[1] nonlinear reflection of ultrashort optical pulses,[2] and "two-wave" solitons[3] which can be treated analytically. These phenomena are typical of exactly integrable Hamiltonians[1–3] and yield important examples of solitons in optics. A model of a thin film of resonant atoms makes it possible to deal also with another class of nonlinear optical phenomena associated with nontrivial dynamics of open dissipative systems. The most familiar phenomena of this kind are optical bistability and spontaneous pulsations (self-pulsations),[4] which are being studied intensively at present because of potential applications in optical data processing and construction of optical computers. Bistability in a film of this kind has been discussed earlier[5] on the basis of a quantum model and more recently[6] using a semiclassical model.

We shall use the semiclassical approach to show that the reflection of a monochromatic light wave by a thin film of two-level atoms as a function of the conditions demonstrates bistability and periodic pulsations. We shall consider a situation in which a thin film is backed by a reflecting surface, for example, the free surface of an insulating substrate which returns part of the transmitted radiation back to the film. A system of this kind exhibits two feedback mechanisms. The first of them is due to the specific nature of the interaction of light with a thin film of resonant atoms and represents a fluctuation-induced reduction in the reflection of light which reduces (because of saturation) the absorption in the film and this in turn reduces the effective refractive index of the substrate and the reflection coefficient of the film-substrate interface. When the surface density of two-level atoms is sufficiently high (in excess of a certain critical value) and for certain intensities of the incident light the same light wave may be reflected in two different steady-state regimes characterized by high and low transparencies of a thin film. The situation is fully analogous to optical bistability of passive low-Q Fabry-Perot resonators filled with resonant atoms if they are considered in the mean field approximation.[7] However, in contrast to the case of optical resonators, when results of this kind are not always correct,[8] the conclusions in the case of a thin film are exact. The second feedback mechanism involves return of the transmitted signal to the film. The phenomena due to this mechanism are largely governed by the phase shift (advance) of the returned signal. If this phase shift is a multiple of 2π, the process of reflection of a monochromatic wave is bistable and, depending on the fraction of the returned signal, the critical value of the surface density of two-level atoms decreases. When the phase shift differs from a multiple of 2π by π, i.e., if the returned signal is in antiphase with the incident one, then suitably modulated transmitted radiation can be compensated fully by the returning radiation so that the reflected radiation may exhibit periodic pulsations and a characteristic hysteresis in steady-state and pulsating regimes. This role of the explicit feedback mechanism can be followed readily by replacing the basic differential-difference equations with point mapping that can be dealt with analytically. In the specific case of the absence of a reflecting surface the expressions obtained become identical with those in Ref. 6.

1. BASIC EQUATIONS

We shall assume that a film of two-level atoms, evaporated on an insulating substrate of thickness L, is illuminated normally by a plane electromagnetic wave and the electric field of this wave is

$$E=\mathscr{E}_I \exp[i(kz-\omega t)]+\text{c.c.},\quad z<0.$$

The wavelength is much greater than the thickness of the film and we shall assume that the film is oriented in the $z=0$ plane. The equations describing the interaction of light with such a film have been discussed repeatedly in the literature.[2] If we allow for relaxation and for the wave reflected from the $z=L$ surface of the insulating substrate, we can rewrite these equations in the following dimensionless form:

$$\left(\frac{d}{d\tau}-i\Delta+\gamma_0\right)p(\tau)=i\varepsilon(\tau)n(\tau), \tag{1}$$

$$\left(\frac{d}{d\tau}+\gamma\right)n(\tau)=2i\{\varepsilon^*(\tau)p(\tau)-\varepsilon(\tau)p^*(\tau)\}+\gamma, \tag{2}$$

$$\varepsilon(\tau)=\psi(\tau)+Re^{i\theta}\psi(\tau-\tau_0), \tag{3}$$

$$p(\tau)=-i\{\psi(\tau)-a(\tau)-\nu Re^{i\theta}\psi(\tau-\tau_0)\}, \tag{4}$$

where

$$n=(\rho_{11}-\rho_{22})N_0^{-1},\quad p=\rho_{21}d^*|d|^{-1}N_0^{-1}\exp(i\omega t),$$

$$a(\tau)=2\mathscr{E}_I|_{z=0}\{(1+n_D)\varepsilon_0\}^{-1},$$

$$\varepsilon(\tau)=(\mathscr{E}_I+\mathscr{E}_R)|_{z=0}\varepsilon_0^{-1}=(\mathscr{E}_F+\mathscr{E}_B)|_{z=0}\varepsilon_0^{-1},$$

$$\psi(\tau)=\mathscr{E}_F|_{z=0}\varepsilon_0^{-1},\ \nu=(n_i-1)(n_i+1)^{-1},\ \tau_0=2Ln_i/ct_0,$$

$$\varepsilon_0=\hbar/t_0|d|,\quad t_0=(1+n_i)\hbar c\{4\pi\omega N_0|d|^2\}^{-1},\quad \tau=t/t_0.$$

The density matrices ρ_{11} and ρ_{22} described the state of an atom at its lower and upper energy levels; ρ_{21} represents a transition between these levels due to the resonant interaction; d is the reduced dipole moment of the transition; N_0 is the steady-state difference between the populations of the

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upper and lower atomic levels in the absence of external fields; n_{Di} is the refractive index of the insulating substrate; R is the reflection coefficient of the $z = L$ surface deduced allowing for the absorption of the signal in the region occupied by the film ($0 < z < L$). Here, Δ, γ_0 and γ represent the dimensionless resonance defect and the rates of relaxation of the polarization and of the difference between the populations of the two-level atoms. The function $\varepsilon(\tau)$ describes the dimensionless amplitude of the electric field in the film, which consists of the incident $\mathscr{E}_I$ and reflected $\mathscr{E}_R$ waves or, which is equivalent, of the transmitted (forward) $\mathscr{E}_F$ and the returned (backward) $\mathscr{E}_B$ waves; $a(\tau)$ is proportional to the amplitude of the incident wave. The quantity τ_0 is the dimensionless time taken by the signal to travel from the $z = 0$ to the $z = L$ plane and back again, whereas s represents the phase shift due to such motion, including a contribution of a possible change due to reflection by the $z = L$ surface.

It is important to stress the following circumstance. If $R = 0$, Eqs. (1)–(4) become identical with the equations given in Ref. 7 and describing, in the mean field approximation, the transmission of a plane light wave through a low-Q Fabry-Perot resonator containing resonant atoms. Therefore, if $R = 0$, then in spite of the difference between the positive feedback mechanisms, reflection of a monochromatic wave by a thin film of resonant atoms may be bistable. At the exact resonance $\Delta = 0$ the cooperation parameter[7] corresponds to

$$C_0 = 1/2\gamma_0 = 2\pi\omega N_0 |d|^2/(1+n_D)\hbar c\Gamma_0. \tag{5}$$

If $C_0 > 4$, then absorption bistability is observed. In Eq. (5) we use $\Gamma_0 = \gamma_0/t_0$ to denote the homogeneous width of a spectral line.

If $R \neq 0$, Eqs. (1)–(4) represent an extremely complex system of equations which cannot be investigated in its general form. However, if the rates γ and γ_0 of the relaxation processes are sufficiently high,

$$\gamma \gg 1/\tau_0, \quad \gamma_0 \gg 1/\tau_0, \tag{6}$$

so that during the time τ_0 of action of the feedback mechanism all the relaxation processes in the thin film decay, i.e., the system becomes of the instantaneous-response type, and all the other changes in time occur on the scale of τ_0, then the derivatives in Eqs. (1) and (2) can be ignored. We can then reduce Eqs. (1) and (2) to their singular limits and the quantities

$$\psi_n = \psi(n\tau_0), \quad a_n = a(n\tau_0), \quad n = 0, 1, 2, \ldots$$

are described by recurrence relationships deduced from the expression

$$a_{n+1} = \psi_{n+1} - \nu R e^{is}\psi_n + \frac{(\gamma_0 + i\Delta)(\psi_{n+1} + Re^{is}\psi_n)}{\gamma_0^2 + \Delta^2 + 4|\psi_{n+1} + Re^{is}\psi_n|^2\gamma_0/\gamma}. \tag{7}$$

The implicit form (7) of point mapping distinguishes the model under consideration from other models of optical systems that can be reduced to mapping (see Ref. 9 and the review in Ref. 10). For simplicity, in the present paper we shall consider only the pure absorption case characterized by $\Delta = 0$, a monochromatic incident wave $a_n = a = \text{const}$, and two values $s = 2\pi m$ and $s = 2\pi(m - 1/2)$, of the phase shift, where $m = 1, 2, \ldots$. Then, Eq. (7) expressed in terms of variables $x_n = 2\psi_n/(\gamma\gamma_0)^{1/2}$ and a parameter $\alpha = 2a/(\gamma\gamma_0)^{1/2}$ becomes

$$\alpha = x_{n+1} - \nu R x_n + \frac{\gamma_0^{-1}(x_{n+1} + Rx_n)}{1 + (x_{n+1} + Rx_n)^2}, \tag{8}$$

where $R > 0$ corresponds to a phase shift amounting to $s = 2\pi m$ and $R < 0 - s = 2\pi(m - 1/2)$.

The different regimes of reflection of a monochromatic wave by a thin film of resonant atoms under conditions defined by Eq. (6) are closely related to stable fixed points and cycles of mapping represented by Eqs. (7) and (8) (Ref. 10). Many relationships governing the appearance of stable cycles of one-dimensional mappings $x_{n+1} = f(x)$ are determined by the presence and positions of extrema of the function $f(x)$ (Ref. 11). In the simplest case when $\nu = 0$ we find that the extremal points $x_e f'(x_e) = 0$ of the mapping of Eq. (8) are as follows:

$$x_e^{(1)} = R^{-1}[1 - \alpha + (2\gamma_0)^{-1}],$$

$$f(x_e^{(1)}) = \alpha - (2\gamma_0)^{-1}, \quad f''(x_e^{(1)}) = R^2(2\gamma_0)^{-1},$$

$$x_e^{(2)} = -R^{-1}[1 + \alpha + (2\gamma_0)^{-1}],$$

$$f(x_e^{(2)}) = \alpha + (2\gamma_0)^{-1}, \quad f''(x_e^{(2)}) = -R^2(2\gamma_0)^{-1}.$$

An analysis of the above shows that in the case of the mapping described by Eq. (8) when $\nu = 0$, there are no regions of stochasticity or sequences of period-doubling bifurcations.

2. OPTICAL BISTABILITY

The fixed points $\bar{x} = f(\bar{x})$ of the mapping of Eq. (8) satisfy the following equation:

$$\alpha = \bar{x}(1 - \nu R) + \frac{(1+R)\gamma_0^{-1}\bar{x}}{1 + (1+R)^2\bar{x}^2}. \tag{9}$$

Using the notation

$$Y = \alpha(1+R)/(1-\nu R), \quad X = (1+R)\bar{x},$$

$$C = (1+R)\{2\gamma_0(1-\nu R)\}^{-1}$$

we find that Eq. (9) reduces to the familiar expression $Y = X(1 + 2C/(1 + X^2))$ of the mean field theory of passive optical cavities containing resonant atoms.[7] If $C > 4$, i.e., when

$$\frac{1+R}{1-\nu R}\,\frac{2\pi\omega N_0|d|^2}{(1+n_i)\hbar c\Gamma_0} > 4, \tag{10}$$

a given value of the amplitude of the incident wave from the interval

$$Y_+ < Y < Y_-, \qquad Y_\pm = \frac{[C - 1 \pm (C^2 - 4C)^{1/2}]^{1/2}}{C \pm (C^2 - 4C)^{1/2}}[3C \pm (C^2 - 4C)^{1/2}] \tag{11}$$

corresponds to three fixed points of the mapping (8). We shall denote them by x_L, x_M, and x_H in accordance with their values $x_L < x_M < x_H$. We can easily show that

$$\left.\frac{d\alpha}{d\bar{x}}\right|_{\bar{x}=x_L} > 0, \quad \left.\frac{d\alpha}{d\bar{x}}\right|_{\bar{x}=x_M} < 0, \quad \left.\frac{d\alpha}{d\bar{x}}\right|_{\bar{x}=x_H} > 0,$$

so that for the same value of α we have

$$\left.\frac{d\alpha}{d\bar{x}}\right|_{\bar{x}=x_L} > -\left.\frac{d\alpha}{d\bar{x}}\right|_{\bar{x}=x_M}. \tag{12}$$

Everywhere with the exception of a narrow ($\sim C^{-1}$ when $C \gg 1$) range of the parameters Y near Y_-, we also have the inequality

$$-\left.\frac{d\alpha}{d\bar{x}}\right|_{\bar{x}=x_M} \geqslant \left.\frac{d\alpha}{d\bar{x}}\right|_{\bar{x}=x_H}. \tag{13}$$

The stability of these fixed points can be determined by considering small deviations $\chi_n = x_n - \bar{x}$. Linearization of Eq. (8) in respect of these deviations gives

$$\chi_{n+1}=R\frac{\nu-\theta}{1+\theta}\chi_n, \tag{14}$$

where

$$\theta=\gamma_0^{-1}\frac{1-\bar{x}^2(1+R)^2}{[1+\bar{x}^2(1+R)^2]^2}.$$

We can easily show that the instability condition of the fixed points $|R(\nu-\theta)/(1+\theta)|>1$ is equivalent to the inequality $d\alpha/d\bar{x}<0$. Therefore, in the region of Eqs. (10) and (11) we have bistable reflection of a monochromatic wave and the dependence of the reflected wave on the incident one is characterized by a hysteresis (Fig. 1a). Introduction of an explicit feedback mechanism (involving reflection from the $z=L$ plane) reduces the critical density of resonant atoms, necessary for bistability reflection of a light wave from a thin film, by a factor $(1+R)/(1-\nu R)$ if $R>0$ and increases this density by a factor $(1+\nu|R|)/(1-|R|)$ if $R<0$.

3. SELF-PULSATIONS

Outside the range defined by Eq. (10), the reflection of a monochromatic wave is a steady-state process if $R>0$ and the dependence of the intensity of the reflected wave on the intensity of the incident wave is single-valued. The situation is different from $R<0$. An analysis is carried out most easily for the values of R close to -1. We shall consider only this case characterized by $R\to -1$. We then have $\bar{x}\simeq\alpha(1+\nu)^{-1}$. It follows from Eq. (14) that in the interval

$$\alpha_-<\alpha<\alpha_+, \qquad \alpha_\pm=\frac{1+\nu}{1-|R|}\left[C_1-1\pm(C_1^2-4C_1)^{1/2}\right]^{1/2}$$

when

$$C_1=\frac{1}{(1-\nu)\gamma_0}=\frac{2}{1-\nu}\frac{2\pi\omega N_0|d|^2}{(1+n_i)\hbar c\Gamma_0}>4 \tag{15}$$

the only fixed point of the mapping (8) becomes unstable. We shall now determine what is the corresponding situation. We shall consider fixed points $\bar{x}_1$ and $\bar{x}_2$ of double iteration of the mapping (8):

$$\alpha=\bar{x}_2-\nu R\bar{x}_1+\frac{\gamma_0^{-1}(\bar{x}_2+R\bar{x}_1)}{1+(\bar{x}_2+R\bar{x}_1)^2},$$

$$\alpha=\bar{x}_1-\nu R\bar{x}_2+\frac{\gamma_0^{-1}(\bar{x}_1+R\bar{x}_2)}{1+(\bar{x}_1+R\bar{x}_2)^2}.$$

If we introduce the notation $\bar{y}_1=(\bar{x}_1+R\bar{x}_2)/(1+|R|)$ and $\bar{y}_2=(\bar{x}_2+R\bar{x}_1)/(1+|R|)$, we find that

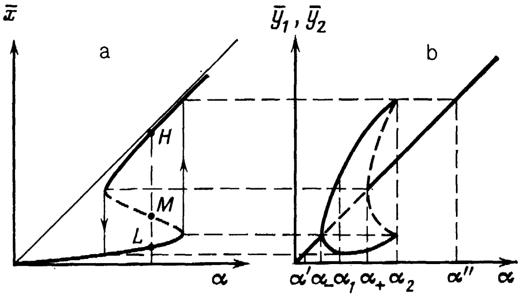

FIG. 1. Dependences of fixed points (a) and 2-cycle points (b) of the mapping described by Eq. (8) on the amplitude α of the incident wave. The dashed curves represent stable points a) This map describes the region defined by Eq. (10). The unit intervals along the abscissa and the ordinate are the quantities $(1+R)/(1-\nu R)$ and $1+R$, respectively. b) Case corresponding to $R\to -1$ subject to the condition (15). The points $\bar{x}_1$ and $\bar{x}_2$ of a 2-cycle of the mapping described by Eq. (8) are related to y_1 and y_2 by $\bar{x}_1+(\bar{y}_1+|R|\bar{y}_2)/(1-|R|)$, and $\bar{x}_2=(\bar{y}_2+|R|\bar{y}_1)/(1-|R|)$. The unit intervals along the abscissa and the ordinate correspond to $(1-|R|)/(1+\nu)$ and 2. The horizontal dashed lines established the correspondence between the parameters of the regime of self-pulsations when $R\to -1$ and the regime of bistability with $R\to 1$; in this case the quantities β and $\bar{y}$ correspond to the abscissa and ordinate in Fig. 1a.

$$(1-\nu|R|)\bar{y}_1+\frac{\gamma_0^{-1}(1+|R|)\bar{y}_1}{1+\bar{y}_1^2(1+|R|)^2}$$

$$=(1-\nu|R|)\bar{y}_2+\frac{\gamma_0^{-1}(1+|R|)\bar{y}_2}{1+\bar{y}_2^2(1+|R|)^2},$$

and in the limit $R\to -1$ we have

$$\bar{y}_1+\bar{y}_2=\alpha(1-|R|)(1+\nu)^{-1}.$$

The solution of these equations is identical with the fixed points of the mapping (8):

$$\beta=y_{n+1}-\nu|R|y_n+\frac{\gamma_0^{-1}(y_{n+1}+|R|y_n)}{1+(y_{n+1}+|R|y_n)^2},$$

which describes bistable reflection of a wave of a certain amplitude β from the same film of resonant atoms, but in this case the phase shift of the returned signal is a multiple of 2π. The value of β is governed by the condition (16). It follows that double iteration of the mapping (8) in the limit $R\to -1$ in the range defined by Eq. (15) is characterized by a nontrivial fixed point forming a 2-cycle $(\bar{y}_1, \bar{y}_2)$ of the original mapping. The following variants of the 2-cycles are possible: (y_L, y_M), (y_L, y_H), and (y_M, y_M). We can determine the range of the parameter α, where this or other 2-cycle exists (Fig. 1b) using the inequalities (12) and (13). For example, in the interval

$$\min(y_L+y_M)\leqslant y_L+y_M\leqslant\max(y_L+y_M)$$

the quantities y_L, y_M, y_L+y_M depend continuously and monotonically on β and Eqs. (12) and (13) are satisfied. Therefore, each parameter α from the interval

$$\alpha_-=\min\frac{(y_L+y_M)(1+\nu)}{(1-|R|)}\leqslant\alpha\leqslant\max\frac{(y_L+y_M)(1+\nu)}{(1-|R|)}$$

$$\equiv\alpha_1=\frac{\alpha_++\alpha'}{2}$$

corresponds to one and just one pair (y_1, y_2), namely $(y_L,$

y_M) such that $y_L + y_M = \alpha(1-|R|)(1+\nu)^{-1}$. At the point $\alpha = \alpha_1$ this 2-cycle transforms continuously to the 2-cycle composed of the points (y_L, y_H):

$$y_L + y_H = \alpha(1-|R|)(1+\nu)^{-1},$$

which is defined in the interval

$$\alpha_1 \leqslant \alpha \leqslant \alpha_2 = \max \frac{(y_H+y_L)(1+\nu)}{2(1-|R|)} \approx \frac{\alpha''+\alpha_-}{2}.$$

At the same time in the range $\alpha_+ \leqslant \alpha \leqslant \alpha_2$ there is a 2-cycle of points (y_H, y_M). It should be stressed that the values of y_H in each pair (y_H, y_L) and (y_H, y_M) are different, since they correspond to different values of the parameter β. It is worth noting the range $\alpha_- \leqslant \alpha \leqslant \alpha_2$ of existence of a 2-cycle of the original mapping (8): it is much wider than the range $\alpha_- \leqslant \alpha \leqslant \alpha_+$ of instability of the fixed point of the same mapping (Fig. 1b).

Physical conclusions can be drawn from the results obtained provided we know the stability of the 2-cycles. We can easily show that the 2-cycles composed of (y_L, y_M) and (y_L, y_H) are stable and the 2-cycle (y_M, y_H) is unstable.

Thus, in absence of a steady-state regime, $\alpha_- \leqslant \alpha \leqslant \alpha_+$ the only stable regime of reflection of a monochromatic wave by a thin film characterized by $R \to -1$ is that of self-pulsations with a period $2\tau_0$ and an amplitude, according to Eq. (3), proportional to $|\bar{y}_1 - \bar{y}_2|$. At the point $\alpha = \alpha_-$ the regimes lose the stability in a soft manner, whereas at the points $\alpha = \alpha_+$ and $\alpha = \alpha_2$ the change in the reflection regime is of the hard type, and in the range $\alpha_+ \leqslant \alpha \leqslant \alpha_2$ there is a characteristic hysteresis of the steady-state pulsating regimes.

4. CONCLUSIONS

The investigated regimes of reflection of a monochromatic light wave from a thin film of resonant atoms are very similar to the passage of a monochromatic wave across a nonlinear optical cavity.[7,9] In a sense, we can even speak of equivalence of optical cavity systems and sets of thin films of two-level atoms and reflecting surfaces. This circumstance widens the range of validity of the results of numerous investigations of lasing and transmission of light across nonlinear optical cavities. On the other hand, it provides a new formulation of the problem of simulation of the transmission of light across nonlinear cavities in which real optical systems are replaced by sets of thin films and mirrors. Since such problems are correctly formulated and—as shown above—they have simple analytic solutions in the ranges defined by Eq. (6), a promising approach is a further study of the interaction of light with other sets of thin films and reflecting surfaces. Moreover, this correspondence between a thin film and a low-Q cavity with resonant atoms, considered in the mean field approximation, makes it possible to apply the results of Ref. 2 to the case of transmission of ultrashort optical pulses by nonlinear cavities and the results of the present study can be applied to transmission of a monochromatic wave across nonlinear resonators with an external mirror.

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[1] V. M. Agranovich, V. I. Rupasov, and V. Ya. Chernyak, Pis'ma Zh. Eksp. Teor. Fiz. **33**, 196 (1981) [JETP Lett. **33**, 185 (1981)].

[2] V. I. Rupasov and V. I. Yudson, Zh. Eksp. Teor. Fiz. **93**, 494 (1987) [Sov. Phys. JETP **66**, 282 (1987)].

[3] B. I. Mantsyzov and R. N. Kuz'min, Zh. Eksp. Teor. Fiz. **91**, 65 (1986) [Sov. Phys. JETP **64**, 37 (1986)].

[4] E. Abraham and S. D. Smith, Rep. Prog. Phys. **45**, 815 (1982).

[5] Y. Ben-Aryeh, C. M. Bowden, and J. C. Englund, Phys. Rev. A **344**, 3917 (1986).

[6] S. M. Zakharov and É. A. Manykin, Poverkhnost' No. 2, 137 (1988).

[7] R. Bonifacio and L. A. Lugiato, Phys. Rev. A **18**, 1129 (1978).

[8] M. Gronchi, V. Benza, L. A. Lugiato, P. Meystre, and M. Sargent III, Phys. Rev. A **24**, 1419 (1981).

[9] H. J. Carmichael, Phys. Rev. Lett. **52**, 1292 (1984).

[10] A. M. Basharov, Photonics: Self-Pulsations and Chaos in Optical Systems [in Russian], Engineering-Physics Institute, Moscow (1987), p.60.

[11] P. Collet and J. P. Eckmann, *Iterated Maps on the Interval as Dynamical Systems*, Birkhauser, Boston, Cambridge, MA (1980), p.248.

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