

Atomic motion in resonant radiation: An application of Ehrenfest's theorem

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A new theory of atomic motion in a resonant or near-resonant electromagnetic wave, based on Ehrenfest's theorem and the optical Bloch equations, is presented. The theory provides a simple unified treatment of the radiation force, including effects of spontaneous emission and induced-dipole interactions. Analytical results are presented for a plane running wave, a general standing wave, a collimated Gaussian beam, and a combination of standing and running waves.

I. INTRODUCTION

The recent revival of interest in the theory of atomic motion in an electromagnetic wave¹⁻⁷ and the increasing number of proposals for the practical application of the theory⁷⁻¹¹ suggest that this subject has a bright future in both pure and applied physics.

Existing theories of atomic motion in resonant radiation tend to fall into one of two categories. There are elementary theories, based largely on intuition and on primitive concepts such as cross section and polarizability, that yield simple formulas for the radiation force under various circumstances, and provide clear physical pictures of the processes involved. These theories tend to be fragmented, with different arguments being used in the derivation of different aspects of the radiation force. For example, the radiation force associated with spontaneous emission and the radiation force associated with interaction of the induced atomic-dipole moment with the amplitude gradient of the applied field are treated separately, and yet another argument is used in discussions of cooling of an atomic vapor by a standing wave tuned below resonance. Such fragmented arguments leave one with the uneasy feeling that perhaps some component of the total radiation force has been neglected, or that an interaction between the different effects might alter the results. On the other hand, there are theories that approach the atom-field interaction from first principles, with both the internal and translational motion of the atom treated quantum mechanically, and often including interaction with the quantized electromagnetic field to take proper account of spontaneous emission. These theories tend to be rather cumbersome, and often numerical calculations must be carried out to obtain useful results.

The purpose of this paper is to present a new approach to the theory of atomic motion in a resonant or near-resonant electromagnetic wave that may be classified approximately midway between the above two categories. The theory, based on Ehrenfest's

theorem and the optical Bloch equations, gives a unified treatment of the radiation force including effects of spontaneous emission and the induced-dipole interaction, and, at the same time, retains much of the simplicity of previous elementary theories.

In Sec. II the theory is developed. In Sec. III explicit formulas are derived for the radiation force in a plane running wave, a standing wave, a collimated Gaussian beam, and a combination of standing and running waves. The paper concludes in Sec. IV with some comments on limitations of the theory.

II. BASIC THEORY

The Hamiltonian for an atom in a classically prescribed electromagnetic wave, in the electric-dipole approximation, is

$$H = P^2/2M + H_0 - \vec{\mu} \cdot \vec{E}(\vec{R}, t), \quad (1)$$

where $P^2/2M$ is the kinetic energy associated with the center-of-mass momentum \vec{P} , H_0 is the Hamiltonian for the internal motion of the unperturbed atom, $\vec{\mu}$ is the electric dipole moment operator, and $\vec{E}(\vec{R}, t)$ is the electric field evaluated at the center-of-mass position \vec{R} . In the Heisenberg representation, operators \vec{R} and \vec{P} satisfy equations of motion

$$\dot{\vec{R}} = (i\hbar)^{-1} [\vec{R}, H] = \nabla_{\vec{P}} H = \vec{P}/M \quad (2)$$

and

$$\dot{\vec{P}} = (i\hbar)^{-1} [\vec{P}, H] = -\nabla_{\vec{R}} H = \nabla(\vec{\mu} \cdot \vec{E}), \quad (3)$$

respectively. Upon combining the expectation values of Eqs. (2) and (3), and setting $\vec{r} = \langle \vec{R} \rangle$, we obtain Ehrenfest's theorem

$$\vec{F} = M \ddot{\vec{r}} = \langle \nabla(\vec{\mu} \cdot \vec{E}) \rangle. \quad (4)$$

To simplify the following calculation, we consider atomic motion in an electric field of the form $\vec{E}(\vec{x}, t) = \hat{e} \mathcal{E}(\vec{x}, t)$, with polarization vector \hat{e} independent of \vec{x} and t . In this case, Eq. (4) becomes \vec{F}

$= \langle \vec{\mu} \cdot \hat{\epsilon} \nabla \mathcal{G} \rangle$, and if $\nabla \mathcal{G}$ is nearly uniform across the atomic wave packet, then

$$\vec{F} = \langle \vec{\mu} \cdot \hat{\epsilon} \rangle \nabla \mathcal{G}(\vec{r}, t). \quad (5)$$

To the same approximation, the internal motion of the atom is driven by the electric vector $\vec{E}(t) = \hat{\epsilon} \mathcal{G}(\vec{r}(t), t)$ at the position of the moving atom, and the Hamiltonian for the internal motion is $H' = H_0 - \vec{\mu} \cdot \vec{E}(t)$.

Now consider the motion of a two-level atom, with energy levels E_1 and E_2 , in an arbitrary monochromatic field

$$\mathcal{G}(\vec{x}, t) = \frac{1}{2} E(\vec{x}) \exp\{i[\theta(\vec{x}) + \omega t]\} + \text{c.c.} \quad (6)$$

Let C_1 and C_2 be the amplitudes that the atom is in levels E_1 and E_2 , respectively. The Schrödinger equation for the internal motion of the atom is then

$$i\hbar \dot{C}_1 = E_1 C_1 - \mu \mathcal{G}(t) C_2, \quad (7)$$

$$i\hbar \dot{C}_2 = E_2 C_2 - \mu \mathcal{G}(t) C_1,$$

where $\mu = \langle 1 | \vec{\mu} \cdot \hat{\epsilon} | 2 \rangle$ is the transition dipole moment (taken here to be real), and $\mathcal{G}(t) = \mathcal{G}(\vec{r}(t), t)$. Upon substituting the relations

$$C_1 = D_1 \exp\{-iE_1 t/\hbar + \frac{1}{2} i[\Delta t + \theta(t)]\}, \quad (8)$$

$$C_2 = D_2 \exp\{-iE_2 t/\hbar - \frac{1}{2} i[\Delta t + \theta(t)]\}$$

into Eqs. (7), with

$$\Delta = \omega - \omega_0 \quad [\omega_0 = (E_2 - E_1)/\hbar]$$

and $\theta(t) = \theta(\vec{r}(t))$, Eqs. (7) become

$$i\hbar \dot{D}_1 = \frac{1}{2} \hbar (\Delta + \dot{\theta}) D_1 - \mu \mathcal{G} D_2 \exp[-i(\theta + \omega t)], \quad (9)$$

$$i\hbar \dot{D}_2 = -\frac{1}{2} \hbar (\Delta + \dot{\theta}) D_2 - \mu \mathcal{G} D_1 \exp[i(\theta + \omega t)].$$

Then inserting (6) into Eqs. (9) and neglecting non-essential terms that oscillate at twice the optical frequency (rotating-wave approximation),¹² Eqs. (9) reduce to

$$i\hbar \dot{D}_1 = \frac{1}{2} \hbar [\Delta + \dot{\theta}(t)] D_1 - \frac{1}{2} \mu E(t) D_2, \quad (10)$$

$$i\hbar \dot{D}_2 = -\frac{1}{2} \hbar [\Delta + \dot{\theta}(t)] D_2 - \frac{1}{2} \mu E(t) D_1.$$

Note that $E(t)$ and $\dot{\theta}(t)$ are determined by the atomic position and velocity through the relations $E = E(\vec{r})$ and $\dot{\theta} = \nabla \theta(\vec{r}) \cdot \dot{\vec{r}}$.

According to Eqs. (8), the density matrix for the internal motion, $\rho_{nm} = C_n C_m^*$, can be written

$$\rho_{11} = \sigma_{11}, \quad \rho_{22} = \sigma_{22},$$

$$\rho_{12} = \sigma_{12} \exp[i(\theta + \omega t)], \quad (11)$$

$$\rho_{21} = \sigma_{21} \exp[-i(\theta + \omega t)],$$

where $\sigma_{nm} = D_n D_m^*$, and it follows from Eqs. (10) that σ_{nm} satisfy equations of motion

$$\dot{\sigma}_{11} = -\frac{1}{2} i \Omega (\sigma_{12} - \sigma_{21}),$$

$$\dot{\sigma}_{22} = \frac{1}{2} i \Omega (\sigma_{12} - \sigma_{21}), \quad (12)$$

$$\dot{\sigma}_{12} = -i(\Delta + \dot{\theta}) \sigma_{12} + \frac{1}{2} i \Omega (\sigma_{22} - \sigma_{11}),$$

where $\Omega(t) = \mu E(t)/\hbar$. $\Omega = \mu E/\hbar$ is the on-resonance Rabi flopping frequency for a two-level atom in a field of amplitude E .

The expectation value appearing in Eq. (5) is written in terms of the density matrix, or in terms of σ_{nm} , as

$$\langle \vec{\mu} \cdot \hat{\epsilon} \rangle = \mu (\rho_{12} + \rho_{21}) = \mu \{ \sigma_{12} \exp[i(\theta + \omega t)] + \sigma_{21} \exp[-i(\theta + \omega t)] \}. \quad (13)$$

Here σ_{nm} are slowly varying functions of time compared to the optical factor $\exp(i\omega t)$. The equation of motion for the atom is obtained by substituting (6) and (13) into Eq. (5) and again discarding inessential terms that oscillate at twice the optical frequency. The result is

$$\vec{F} = M \ddot{\vec{r}} = \frac{1}{2} \mu \nabla E (\sigma_{12} + \sigma_{21}) - \frac{1}{2} i \mu E \nabla \theta (\sigma_{12} - \sigma_{21}). \quad (14)$$

Effects of spontaneous emission are introduced into the theory by adding relaxation terms to Eqs. (12).

$$\dot{\sigma}_{11} = -\frac{1}{2} i \Omega (\sigma_{12} - \sigma_{21}) + A \sigma_{22},$$

$$\dot{\sigma}_{22} = \frac{1}{2} i \Omega (\sigma_{12} - \sigma_{21}) - A \sigma_{22}, \quad (15)$$

$$\dot{\sigma}_{12} = -i(\Delta + \dot{\theta}) \sigma_{12} + \frac{1}{2} i \Omega (\sigma_{22} - \sigma_{11}) - \frac{1}{2} A \sigma_{12},$$

where $A = 4\omega_0^3 |\langle 1 | \vec{\mu} | 2 \rangle|^2 / 3\hbar c^3$ is the spontaneous emission rate (Einstein A coefficient). The relaxation terms may be derived from a first-principles calculation¹³ or simply written on the basis of simple phenomenological arguments.

Equations (14) and (15) can be rewritten in terms of three real variables

$$U = (\sigma_{12} + \sigma_{21}), \quad V = -i(\sigma_{12} - \sigma_{21}), \quad W = (\sigma_{22} - \sigma_{11}), \quad (16)$$

as

$$\vec{F} = M \ddot{\vec{r}} = \frac{1}{2} \hbar (U \nabla \Omega + V \Omega \nabla \theta) \quad (17)$$

and

$$\dot{U} = (\Delta + \dot{\theta}) V - \frac{1}{2} A U,$$

$$\dot{V} = -(\Delta + \dot{\theta}) U + \Omega W - \frac{1}{2} A V, \quad (18)$$

$$\dot{W} = -\Omega V - A(W + 1),$$

respectively, where $\sigma_{11} + \sigma_{22} = 1$ was used in the derivation of (18). Equations (18) are the optical Bloch equations in the rotating wave approximation.

Equations (17) and (18) determine the motion of a two-level atom in a monochromatic field with arbitrary amplitude $E(\vec{x})$ and phase $\theta(\vec{x})$. It is clear from these equations that, in general, the radiation force is not a simple function of the atoms position and velocity, but rather depends on the history of the motion through the Bloch equations. There are, however, certain cases in which the radiation force reduces to a function of atomic position and velocity to an excellent approximation. Some of these cases are discussed in Sec. III.

III. EXAMPLES

A. Steady-state approximation

Consider first the case in which the electric field amplitude $E(t) = E(\vec{r}(t))$ and phase derivative $\dot{\theta}(t) = \nabla\theta(\vec{r}(t)) \cdot \dot{\vec{r}}(t)$ vary by only a small fraction during a natural lifetime $\tau_N = 1/A$, i.e., the case in which the atom moves sufficiently slowly so that at each instant U , V , and W assume the steady-state values obtained from Eqs. (18) by setting $\dot{U} = \dot{V} = \dot{W} = 0$. The steady-state solution of Eqs. (18) gives

$$U = -4\Omega(\Delta + \dot{\theta}) / [4(\Delta + \dot{\theta})^2 + A^2 + 2\Omega^2],$$

$$V = -2A\Omega / [4(\Delta + \dot{\theta})^2 + A^2 + 2\Omega^2],$$

and hence, the radiation force, Eq. (17), becomes

$$\vec{F} = -[\hbar A \Omega^2 \nabla\theta + \hbar(\Delta + \dot{\theta}) \nabla\Omega^2] / [4(\Delta + \dot{\theta})^2 + A^2 + 2\Omega^2]. \quad (19)$$

The meaning of Eq. (19) will be illustrated by applying it to a few simple problems.

For a plane running wave, $\mathcal{E}(\vec{x}, t) = \mathcal{E}_0 \cos(\vec{k} \cdot \vec{x} - \omega t)$, we have $\Omega = \mu \mathcal{E}_0 / \hbar = \text{const}$, $\theta(\vec{x}) = -\vec{k} \cdot \vec{x}$, and $\dot{\theta} = -\vec{k} \cdot \dot{\vec{r}}$. The radiation force, Eq. (19), reduces to

$$\vec{F} = A\Omega^2 \hbar \vec{k} / [4(\Delta - \vec{k} \cdot \dot{\vec{r}})^2 + A^2 + 2\Omega^2]. \quad (20)$$

This is the radiation force associated with spontaneous emission or with scattering of radiation by the atom. The force is a Lorentzian function of ω centered at $\omega_0 + \vec{k} \cdot \dot{\vec{r}}$ (atomic frequency plus Doppler shift) with full width at half-maximum $(A^2 + 2\Omega^2)^{1/2}$ corresponding to natural and power broadening of the atomic response. In a strong field ($\Omega \rightarrow \infty$) the force saturates to the value $\vec{F} = \frac{1}{2} A \hbar \vec{k}$. Equation (20) is consistent with Ashkin's theory of resonance-radiation pressure.⁷

In a general standing wave, $\mathcal{E}(\vec{x}, t) = E(\vec{x}) \cos \omega t$, we have $\Omega(\vec{x}) = \mu E(\vec{x}) / \hbar$ and $\theta = 0$. Here the amplitude $E(\vec{x})$ is a solution of the time-independent wave equation $\nabla^2 E + (\omega/c)^2 E = 0$. The radiation force, Eq. (19), is now

$$\vec{F} = -\hbar \Delta \nabla \Omega^2 / (4\Delta^2 + A^2 + 2\Omega^2). \quad (21)$$

This force is a result of the interaction between the induced atomic dipole moment and the amplitude gradient of the standing wave. It may be written in the form $\vec{F} = \frac{1}{2} \alpha \nabla E^2$, where

$$\alpha = -2\Delta \mu^2 / \hbar [4\Delta^2 + A^2 + 2(\mu E / \hbar)^2]$$

is the atomic polarizability. The dipole force is derivable from a potential

$$\vec{F} = -\nabla U, \quad U = \frac{1}{2} \hbar \Delta \ln(4\Delta^2 + A^2 + 2\Omega^2). \quad (22)$$

When the field is tuned above resonance ($\Delta > 0$) the dipole force is in the direction of decreasing field strength, and the atom tends to be expelled by the field. When the field is tuned below resonance

($\Delta < 0$) the dipole force is in the direction of increasing field strength, and the atom tends to be trapped by the radiation. On resonance ($\Delta = 0$) the dipole force vanishes. These results for a general standing wave are again consistent with the theory of Ashkin.⁷

Equation (19) may also be applied to problems involving a combination of standing and running waves. In this case, a new velocity-dependent term appears which has not been considered in previous treatments of radiation force. For example, if the applied field

$$\mathcal{E}(x, t) = \mathcal{E}_s \cos kx \cos \omega t + \mathcal{E}_T \cos(kx - \omega t) \quad (23)$$

is written in the form of Eq. (6), and the resulting amplitude $E(x)$ and phase $\theta(x)$ are inserted into Eq. (19), the radiation force becomes

$$F_x = \frac{A\Omega_1\Omega_2\hbar k}{D} - \frac{\hbar\Delta[\Omega^2]'}{D} + \frac{\hbar\Omega_1\Omega_2 k \dot{x} [\Omega^2]'}{\Omega^2 D}, \quad (24)$$

where

$$\Omega_1 = \mu \mathcal{E}_T / \hbar, \quad \Omega_2 = \mu (\mathcal{E}_T + \mathcal{E}_s) / \hbar,$$

and

$$\Omega^2 = \Omega_1^2 \sin^2 kx + \Omega_2^2 \cos^2 kx,$$

$$D = 4(\Delta + \dot{\theta})^2 + A^2 + 2\Omega^2, \quad (25)$$

$$\dot{\theta} = -\Omega_1 \Omega_2 k \dot{x} / \Omega^2.$$

The first two terms in (24) will be recognized as generalizations of the running and standing wave forces considered above, while the third term is new and occurs only when standing and running waves are simultaneously present. When the atomic velocity is zero, the new term vanishes, and the dipole force in (24) is derivable from the potential (22) with $\Omega^2(x)$ taken from Eq. (25). The depth of modulation of this periodic potential is

$$\delta U = U_{\max} - U_{\min} = \frac{1}{2} \hbar \Delta \ln \left(\frac{4\Delta^2 + A^2 + 2(\Omega_s + \Omega_1)^2}{4\Delta^2 + A^2 + 2\Omega_1^2} \right), \quad (26)$$

where $\Omega_s = \mu \mathcal{E}_s / \hbar$. As the strength of the running wave Ω_1 increases from zero, the trapping energy δU of the standing wave first increases to a maximum and then decrease to zero as $\hbar \Delta (\Omega_s / \Omega_1) = \hbar \Delta (\mathcal{E}_s / \mathcal{E}_1)$. This result disagrees with the theory of Ashkin⁷ which predicts a monotonic decrease of the trapping energy with increasing \mathcal{E}_1 , and a limiting value proportional to $(\mathcal{E}_s / \mathcal{E}_1)^2$ as $\mathcal{E}_1 \rightarrow \infty$.

Another case of current experimental interest is that of atomic motion in a Gaussian laser beam. Consider a collimated Gaussian beam of spot size w_0 propagating in the z direction.

$$\mathcal{E}(\vec{x}, t) = \mathcal{E}_0 \exp[-(x^2 + y^2)/w_0^2] \cos(kz - \omega t). \quad (27)$$

Here

$$\Omega(x, y) = (\mu \mathcal{E}_0 / \hbar) \exp[-(x^2 + y^2)/w_0^2]$$

and $\theta(z) = -kz$. The radiation force, Eq. (19), consists of a longitudinal radiation pressure

$$F_z = A\Omega^2 \hbar k / [4(\Delta - k\dot{z})^2 + A^2 + 2\Omega^2] \quad (28)$$

and a transverse dipole force

$$\vec{F}_T = -\hbar(\Delta - k\dot{z})\nabla\Omega^2 / [4(\Delta - k\dot{z})^2 + A^2 + 2\Omega^2]. \quad (29)$$

Unlike the dipole force in a standing wave, which is independent of velocity (in the present approximation), the transverse-dipole force depends on the atomic velocity through the Doppler shift $k\dot{z}$. The dipole force is directed toward the beam axis when $k\dot{z} > \Delta$ and away from the axis when $k\dot{z} < \Delta$. This result leads to the interesting prediction that in a resonant Gaussian beam ($\Delta = 0$) a copropagating atomic beam ($\dot{z} > 0$) is focused and trapped by the field, while a counterpropagating atomic beam ($\dot{z} < 0$) is defocused and expelled by the resonant radiation. Focusing and defocusing of a beam of sodium atoms by the transverse dipole force in a copropagating Gaussian laser beam has recently been observed in the experiment of Bjorkholm *et al.*¹⁴

B. Dissipative force in a standing wave

When an atom moves with typical thermal velocity v across the fringes of a simple standing wave $E(x) = \mathcal{E}_0 \cos kx$ of visible light, the approximation of slowly varying field amplitude that lead to Eq. (19) is no longer valid [the amplitude at the moving atom varies as $E(t) = \mathcal{E}_0 \cos kvt$ and kv generally exceeds the spontaneous emission rate A]. Thus the above standing-wave results are valid only in the limit $kv \ll A$, and the case $kv \geq A$ requires a different approach. In this subsection we calculate the time-average radiation force for arbitrary atomic velocity in a weak standing wave. This problem is of considerable interest in connection with recent proposals for cooling an atomic vapor by a standing wave tuned below resonance.^{7,9}

In a simple standing wave, $\Omega(x) = \Omega_0 \cos kx$ ($\Omega_0 = \mu \mathcal{E}_0 / \hbar$), $\theta = 0$, and the force acting on the atom, Eq. (17), is $F_x = -\frac{1}{2} U \Omega_0 \hbar k \sin kx$. Let $x = vt$. Then the time-average radiation force is

$$\bar{F}_x = -\frac{1}{2} \Omega_0 \hbar k \langle U(t) \sin kvt \rangle_{av}, \quad (30)$$

and the equations describing the internal motion of the atom, Eqs. (18), are

$$\dot{U} = -\frac{1}{2} AU + \Delta V, \quad (31a)$$

$$\dot{V} = -\frac{1}{2} AV - \Delta U + \Omega_0 \cos(kvt)W, \quad (31b)$$

$$\dot{W} = -\Omega_0 \cos(kvt)V - A(W + 1). \quad (31c)$$

If the field is weak ($\Omega_0 \ll A$), the degree of atomic excitation (or inversion) W remains near the ground-state value ($W \approx -1$). For $W = -1$, Eqs. (31a) and (31b) can be solved exactly. The persistent solution is

$$U = \alpha \cos kvt + \beta \sin kvt, \quad (32)$$

$$V = \gamma \cos kvt + \nu \sin kvt, \quad (33)$$

where

$$\alpha = -\Delta\Omega_0 [(\frac{1}{2}A)^2 + \Delta^2 - (kv)^2] / D, \\ \beta = -\Delta\Omega_0 A kv / D, \quad (34)$$

$$\gamma = -\frac{1}{2} A \Omega_0 [(\frac{1}{2}A)^2 + \Delta^2 + (kv)^2] / D,$$

$$\nu = -\Omega_0 kv [(\frac{1}{2}A)^2 - \Delta^2 + (kv)^2] / D,$$

and

$$D = [\Delta^2 - (kv)^2]^2 \\ + (\frac{1}{2}A)^2 [(\frac{1}{2}A)^2 + 2\Delta^2 + 2(kv)^2]. \quad (35)$$

Insertion of (32) into (30) yields $\bar{F}_x = -\frac{1}{4} \Omega_0 \hbar k \beta$, and taking β from (34), we obtain

$$\bar{F}_x = \frac{v \Delta A \Omega_0^2 \hbar k^2}{4[\Delta^2 - (kv)^2]^2 + A^2 [(\frac{1}{2}A)^2 + 2\Delta^2 + 2(kv)^2]}. \quad (36)$$

This equation states that the time-average radiation force in a weak standing wave is a positive quantity times $v\Delta$. Thus a standing wave tuned below resonance ($\Delta < 0$) damps the atomic velocity, while a standing wave tuned above resonance ($\Delta > 0$) amplifies the atomic velocity.

A reliable calculation of the radiation force in a strong standing wave requires solution of the full set of equations (31). This problem is more difficult than the simple examples considered here and will not be discussed in the present paper.

IV. CONCLUSION

It should be emphasized that a theory based on Ehrenfest's theorem describes the motion of the centroid of the center-of-mass probability density. It says nothing about the spread of the atomic wave packet about the centroid. Because of this limitation, results of the present theory are, in some cases, misleading. For example, the present theory suggests that the radiation force acting on a slowly moving atom in a standing wave vanishes as $\Delta \rightarrow 0$, while a more detailed theory¹⁵ shows that, for $\Delta = 0$, the atomic trajectory is split by the resonant radiation, in much the same way as a narrow atomic beam is split in the Stern-Gerlach experiment, but the splitting is symmetric so the centroid is not accelerated. This lack of detail in the present theory is the price paid for simplicity. On the other hand, in almost all problems of practical interest, the deBroglie wavelength of the atom is

many orders of magnitude smaller than the optical wavelength (the minimum scale size of the applied field), and therefore it is expected that the simple picture of a point atom moving along a classical trajectory is an excellent approximation for a wide class of problems.

The present theory is clearly applicable to more elaborate problems than considered here. We believe that our basic working equations, Eq s. (17) and (18), will provide a convenient and fruitful

framework in which to study such problems.

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