# **Nonlinear Rayleigh waves**

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Self-induced transparency is investigated theoretically for Rayleigh waves interacting with the electron-nuclear spin system of paramagnetic impurities. Explicit solutions are found which correspond to McCall-Hahn and breather  $2\pi$ -solitons. The parameters of the nonlinear Rayleigh waves are shown to depend on the transverse structure of the field. The possibility of detecting the self-induced transparency experimentally is discussed.

## **1. INTRODUCTION**

The phenomenon of acoustic self-induced transparency (SIT) can be used to investigate the properties of acoustic solitons in resonantly absorbing condensed media. The theoretical treatments consider a model in which volume plane waves propagate in an unbounded medium. The problem then reduces to solving a nonlinear system of equations involving time and a single spatial coordinate. This system is exactly solvable by the inverse scattering method,<sup>1</sup> and its solutions were analyzed in detail in Ref. 2. Such a model correctly describes the experimental data on acoustic SIT when the propagation of the acoustic waves is not significantly affected by the presence of solid boundaries.<sup>3,4</sup>

In bounded (multilayer) systems the situation is different, because the interfaces between the layers, which have different elastic properties, can generate surface acoustic waves whose structure differs greatly from that of internal plane waves. The most important case involves Rayleigh waves, which can propagate either near a free solid surface or along the interface between a solid half-space and a solid layer. High-frequency Rayleigh waves are easily produced and are currently the subject of numerous experimental investigations (see, e.g., Ref. 5 and the bibliography cited therein). If the Rayleigh waves travel in a medium containing paramagnetic impurities, the wave-impurity interaction can excite resonant transitions in the impurities. There are two situations of particular interest: 1) the paramagnetic impurities form a thin resonance layer on the solid surface; 2) the entire medium is resonant. In contrast to the case of internal plane waves, the analysis of SIT for Rayleigh waves requires that one solve a nonlinear two-dimensional system of equations (two spatial coordinates plus time) self-consistently together with the evolution equations for a spin system interacting with the Rayleigh wave. No general method is available for doing this analytically, and a detailed numerical analysis lies beyond the capabilities of existing computers. Self-induced transparency for Rayleigh waves interacting with paramagnetic impurities is investigated theoretically in the present paper, where we employ approximate methods to find some explicit solutions (breather and McCall-Hahn  $2\pi$ -solitons).

## 2. DERIVATION OF THE EQUATIONS

The basic features of SIT for Rayleigh waves can be analyzed by considering a simple model in which a nonmetallic diamagnetic solid contains a small number of paramagnetic impurities with electron and nuclear spins S and I. For simplicity we take S = I = 1/2 and assume that the solid medium fills a half-space x < 0. We consider a Rayleigh wave pulse whose duration T is much shorter than the irreversible relaxation times; the wave has wave vector Q and frequency  $\omega_Q$ , and it propagates along the positive z axis on the surface x = 0. A constant magnetic field  $H_0$  is also applied along the z axis. We will analyze the case when the Rayleigh wave excites forbidden transitions in the electron-nuclear spin system (transitions in which both the electron and the nuclear spins change direction). In this case  $\omega_Q \approx \omega_S + \omega_I$ , where  $\omega_S$  and  $\omega_I$  are the Zeeman frequencies for the electron and nuclear spins (see Ref. 6 for the case of internal acoustic plane waves).

The deformation vector  $\mathbf{u}$  in the Rayleigh wave is expressible as  $\mathbf{u}_l + \mathbf{u}_r$ , where  $\nabla \times \mathbf{u}_l = 0$  and  $\nabla \cdot \mathbf{u}_r = 0$  (Ref. 7). In accordance with the above model, we assume that no strain is present on the free surface x = 0:

$$\sigma_{xx} = \sigma_{xy} = \sigma_{xz} = 0. \tag{1}$$

Among the solutions satisfying these boundary conditions we consider those that correspond to Rayleigh waves, whose amplitude decays exponentially inside the solid as  $x \to -\infty$ . Using  $\nabla \times \mathbf{u}_t = \nabla \cdot \mathbf{u}_t = 0$ , we can write

$$u_{z}(x, z, t) = \sum_{k} \beta_{k}(x) \left[ a_{k} e^{ikz} + a_{k}^{+} e^{-ikz} \right] (2\rho \omega_{k})^{-1/2},$$

for the z-component of u of interest, where the quantity

$$\beta_k(x) = e^{\kappa_l(k)x} - 2\kappa_l(k)\kappa_t(k) [\kappa_t^2(k) + k^2]^{-1} e^{\kappa_l(k)x}$$

determines the transverse structure of the field; the creation and annihilation operators  $a_k^+$  and  $a_k$  for the Rayleigh modes satisfy the commutation relations<sup>8,9</sup>

 $[a_{k}, a_{k'}] = [a_{k}^{+}, a_{k'}^{+}] = 0, \quad [a_{k}, a_{k'}^{+}] = \delta_{kk'};$ 

the quantities  $x_i(k)$  and  $x_i(k)$ , which determine the rate of wave damping along the x axis, are given by

$$\varkappa_{l,t}(k) = [k^2 - (\omega_k/c_{l,t})^2]^{1/2}, \qquad (2)$$

where  $c_i$  and  $c_i$  are the longitudinal and transverse wave velocities;  $\rho$  is the density of the medium, and  $\omega_k$  is the frequency for the k th mode.

The boundary condition  $\sigma_{xy}|_{x=0} = 0$  implies that the deformation vector **u** in the Rayleigh wave lies in a plane perpendicular to the surface and containing the z axis, i.e.,  $u_y = 0$ . It follows from the remaining boundary conditions in (1) that  $\omega_k$  and k are related by

$$[\varkappa_{t}^{2}(k)+k^{2}]^{2}=4k^{2}\varkappa_{t}(k)\varkappa_{t}(k).$$
(3)

Comparison of (2) and (3) yields  $\omega_k = ck$ , where c is the velocity of the Rayleigh wave.

In the rotating wave approximation,<sup>10</sup> the Hamiltonian of the system is given by

$$\mathcal{H} = \omega_{s} S^{z} - \omega_{I} I^{z} + A \sum_{i} I_{i}^{z} S_{i}^{z}$$

$$- ig \sum_{i} (S_{i}^{+} I_{i}^{-} A_{i}^{-} - S_{i}^{-} I_{i}^{+} A_{i}^{+}) \qquad (4)$$

$$+ \sum_{i} \omega_{k} (a_{k}^{+} a_{k}^{+} + i/_{2}),$$

where we have set  $\hbar = 1$ . Here  $g = AH_0F_{zzzz}\beta/2\omega_0$ ;  $\beta$  is the Bohr magneton; A is the hyperfine interaction constant;  $\varepsilon_{zz} = \partial u_z / \partial z = i(A^- - A^+)$ ; the  $F_{zzzz}$  are the components of the deformation and spin-phonon coupling tensors; and

$$A_{l}^{\pm} = \sum_{\mathbf{k}} \beta_{\mathbf{k}}(x_{l}) \eta_{\mathbf{k}} a_{\mathbf{k}}^{\pm} \exp(\mp i k z_{l}), \quad \eta_{\mathbf{k}} = (2\rho\omega_{\mathbf{k}})^{-\gamma_{\mathbf{k}}} k.$$

The Hamiltonian (4) leads to the following Heisenberg equations of motion for the spin and field mode operators:

$$i\dot{a}_{k} = \omega_{k}a_{k} + ig \sum_{l} S_{l}^{-}I_{l}^{+}u^{*}(k,\mathbf{r}_{l}),$$

$$(S_{l}^{+}I_{l}^{-})^{*} = i(\omega_{s} + \omega_{l})S_{l}^{+}I_{l}^{-} + g(S_{l}^{z} - I_{l}^{z})A_{l}^{+},$$

$$S_{l}^{z} - I_{l}^{z} = -2g(S_{l}^{+}I_{l}^{-}A_{l}^{-} + S_{l}^{-}I_{l}^{+}A_{l}^{+}),$$
(5)

where

$$u(k, \mathbf{r}_l) = \beta_k(x_l) \eta_k \exp(ikz_l)$$

(we neglect relaxation and phase modulation effects). This system of SIT equations for Rayleigh waves is valid for any distribution of paramagnetic impurities in the half-space *x*≼0.

Further simplification can be achieved by replacing the operator equations (5) by the corresponding equations for the expectation values. In the semiclassical approximation, for which the expectation values for a product of operators of the type  $A \pm S^z$  can be factored as<sup>10</sup>

 $\langle A^{\pm}S^{z}\rangle \approx \langle A^{\pm}\rangle \langle S^{z}\rangle,$ 

we obtain the equation

$$\dot{\alpha}_{k} = -i\omega_{k}\alpha_{k} + g \sum_{l} \mathcal{B}_{l} - u^{*}(k, \mathbf{r}_{l})$$
(6)

for the acoustic field and the following system of equations for the variables  $B_{l}^{\pm}$  and  $N_{l}$ :

$$\dot{B}_{l}^{+} = i(\omega_{s} + \omega_{l} - \omega_{k})B_{l}^{+} + 2gN_{l}a_{l}^{+},$$
  
$$\dot{N}_{l} = -g(B_{l}^{+}a_{l}^{-} + B_{l}^{-}a_{l}^{+}),$$
(7)

where the quantities  $\alpha_k$ ,  $\widetilde{B}_l^{\pm}$ , and  $N_l$  are defined by

 $\cdot a_k |\alpha_k\rangle = \alpha_k |\alpha_k\rangle,$  $A_{l} = \langle A_{l} \rangle = \sum_{k} \beta_{k}(x_{l}) \eta_{k} \alpha_{k} \exp(ikz_{l}) = a_{l} \exp[-i(\omega_{Q}t - Qz_{l})],$  $a_{l} \equiv a(x_{l}, z_{l}, t) = \beta_{Q}(x_{l}) \varepsilon(z_{l}, t) - i(\partial \beta_{k}/\partial k)_{k=Q} [\partial \varepsilon(z_{l}, t)/\partial z_{l}],$  $\langle S_l^{\pm}I_l^{\pm}\rangle = \widetilde{B}_l^{\pm} = B_l^{\pm} \exp[\pm i(\omega_Q t - Qz_l)],$  $2N_l = \langle S_l^i - I_l^i \rangle, \quad B_l^{\pm} = u_l \pm i v_l.$ (8)

Here  $|\alpha_k\rangle$  gives the k th coherent-state mode for the surface phonons.8,9

#### 3. THE MCCALL-HAHN 2π-SOLITON

We consider the case when the paramagnetic impurities are contained in a monolayer at the surface of the medium. The number of active particles per unit volume is then  $N(x_l) = N_0 \delta(x_l)$ , and the quantity  $\tilde{B}_l^-$  is given by  $\tilde{B}_{l}^{-} = \delta(x_{l})\tilde{B}_{1}^{-}(z_{l},t)$ . Since all of the basic properties of SIT can be studied at the precise resonance  $\omega_O = \omega_S + \omega_I$ , we will solve the SIT equations in this case:

$$u_l = -\frac{1}{2} \sin \Psi(z_l, t), \quad v_l = 0, \quad N_l = -\frac{1}{2} \cos \Psi(z_l, t),$$
 (9)

where

$$\widetilde{B}_{1}^{-}(z_{l},t) = (u_{l}-iv_{l}) \exp\left[-i(\omega_{Q}t-Qz_{l})\right],$$

$$\Psi(z_{l},t) = 2g\beta_{Q}(0) \int_{-\infty} \varepsilon(z_{l},t') dt',$$

 $\Psi$  is the area of the envelope of the Rayleigh wave pulse.

Multiplying Eq. (6) by  $\beta_Q(0)\eta_k \exp(ikz_l)$ , summing over k, and using Eq. (8), we obtain

$$\frac{\partial A}{\partial t} = -c \frac{\partial A}{\partial z_{l}} + g\beta_{Q}(0) \sum_{L} \tilde{B}_{1L} \sum_{k} \xi_{k} \exp[ik(z_{l}-z_{L})], \qquad (10)$$

where  $\xi_k = \eta_k^2 \beta_k(0)$ , and L refers to the yz plane.

We expand  $\xi_k$  as a Taylor series about the mean value of the wave vector Q:

$$\xi_k = \xi_Q + \Delta k \left( \partial \xi_k / \partial k \right)_{k=Q} + O\left[ \left( \Delta k / Q \right)^2 \right], \quad \Delta k = k - Q.$$

Substituting this into Eq. (10), discarding terms of order  $(\Delta k/Q)^2$ , and separating real and imaginary parts, we get the relation

$$\omega_Q = cQ,$$

between  $\omega_Q$  and Q and the equation

$$\partial^2 \Psi / \partial t^2 + c \partial^2 \Psi / \partial t \partial z_l = -\alpha_0^2 \sin \Psi, \qquad (11)$$

for the area  $\Psi$  of the envelope; here

$$\alpha_0^2 = \frac{g^2 N_0 \omega_Q}{8\rho c_i^2} \left(\frac{c}{c_i}\right)^2$$

Equation (11), the familiar sine-Gordon equation, is exactly solvable by the inverse scattering method and admits a soliton solution.<sup>1</sup> Equation (11) can be solved more simply by passing to the variable  $\tau = t - z_1/V$ , where V is the velocity of the soliton. Equation (11) then becomes

$$d^2\Psi/d\tau^2 = T^{-2}\sin\Psi,$$

where T is the width of the pulse, and it has the solution

$$a(\tau) = \frac{2}{Tg} \operatorname{ch}^{-1} \frac{\tau}{T}, \quad T^{-2} = \frac{g^2 N_0 Q \beta_Q^2(0)}{2\rho c [c/V - 1]}, \quad (12)$$

which corresponds to a McCall-Hahn  $2\pi$ -pulse (soliton).<sup>10</sup> It is evident from these expressions that unlike the case of internal plane-wave solitons, the parameters of the Rayleigh wave soliton depend on the factor  $\beta_Q(0) = c^2/2c_t^2$ , which allows for the transverse structure of the field.

These results can be generalized as in Ref. 2 to the slightly off-resonance case and to allow for inhomogeneous broadening and relaxation effects, for which the transverse field structure plays no role.

Since  $c = \xi_0 c_t$ , where  $0.87 < \xi_0 < 0.96$  (Ref. 7), it is evident from Eq. (12) that for a given duration *T*, the soliton velocity for a Rayleigh wave is of the same order of magnitude as for internal plane-wave solitons. Moreover, the solutions (12) can be observed experimentally under roughly the same conditions (field and two-level system parameters) as for the internal plane waves studied experimentally in Refs. 3 and 4.

#### 4. PULSATING SOLITON (BREATHER)

We now consider the case when the paramagnetic impurities are distributed uniformly throughout a bulk solid medium. To solve the system (6), (7) at the precise resonance  $\omega_Q = \omega_S + \omega_I$ , we recall that  $(\partial \beta_k / \partial k)_{k=Q} \sim Q^{-1}$ and obtain

$$u = -\frac{n}{2}\sin\Psi_{i} + O\left(\frac{1}{\varepsilon Q}\frac{\partial\varepsilon}{\partial z}\right), \quad v = O\left(\frac{1}{\varepsilon Q}\frac{\partial\varepsilon}{\partial z}\right),$$
$$N = -\frac{n}{2}\cos\Psi_{i} + O\left(\frac{1}{\varepsilon Q}\frac{\partial\varepsilon}{\partial z}\right), \quad (13)$$

where

$$\Psi_{i}(x,z,t) = 2\beta_{Q}(x)g\int_{-\infty}^{\infty} \varepsilon(z,t')dt'$$

is the area of the envelope of the Rayleigh wave and n is the number of active particles per unit volume. We note that in contrast to the previous case,  $\Psi_1$  also depends on the transverse coordinate x.

Equations (13) can be solved if all the amplitudes are assumed to vary only slowly with z and t:

$$\left| \frac{\partial \varepsilon}{\partial t} \right| \ll \omega_{Q} |\varepsilon|, \qquad \left| \frac{\partial \varepsilon}{\partial z} \right| \ll Q |\varepsilon|,$$
$$\left| \frac{\partial u}{\partial z} \right| \ll Q |u|, \qquad \left| \frac{\partial u}{\partial t} \right| \ll \omega_{Q} |u|.$$

Multiplying Eq. (6) by  $\eta_k \exp(ikz_{l'})$ , adding over k, and using (8), we obtain

$$\frac{\partial A_{l'}}{\partial t} = -c \frac{\partial A_{l'}}{\partial z_{l'}} + g \sum_{l} \sum_{k} B_{l} - s_{k}(x_{l}) \exp[ik(z_{l'} - z_{l})]$$

where

$$s_k(x_l) = \eta_k^2 \beta_k(x_l).$$

Expanding  $s_k(x_l)$  as a Taylor series about the average wave vector Q, we obtain the system of equations

$$\omega_{\mathbf{Q}} = cQ,$$
  
$$\frac{\partial^2 \Psi_{\mathbf{i}}'}{\partial t^2} + c \frac{\partial^2 \Psi_{\mathbf{i}}'}{\partial z \partial t} = -R^2 \sum_{x_i} \beta_{\mathbf{Q}}(x_i) \sin[\beta_{\mathbf{Q}}(x_i) \Psi_{\mathbf{i}}'] + O\left(\frac{\Delta k}{Q}\right),$$

after some straightforward algebra, where

$$\Psi_1' = 2g \int_{-\infty}^t \varepsilon(z,t') dt', \quad R^2 = g^2 n Q/2\rho c.$$

The last equation can be solved for small Rayleigh wave amplitudes  $\Psi' \ll 1$ . Using the series expansion of  $\sin(\beta_Q \Psi'_1)$ , we get the following nonlinear equation

$$\frac{\partial^2 \Psi_i}{\partial t^2} + c \frac{\partial^2 \Psi_i}{\partial z \, \partial t} = -\alpha_i^2 \Psi_i' + \alpha_2^2 \Psi_i'^3 - O(\Psi_i'^5) + O\left(\frac{\Delta k}{Q}\right),$$
(14)

where

$$\alpha_1^2 = R^2 \xi_1, \quad \alpha_2^2 = \frac{1}{6} R^2 \xi_2,$$
  
$$\xi_1 = \sum_{x_1} \beta_Q^2(x_1), \quad \xi_2 = \sum_{x_2} \beta_Q^4(x_2).$$

If one seeks a solution of the form

 $\Psi_{i} = \Phi(z, t) \exp[i(\Omega_{0}t - qz)] + \text{c.c.},$ 

Eq. (14) can be solved explicitly as in Refs. 11 by transforming it into the nonlinear Schrödinger equation. In order to do this, the condition  $\Psi'_1 \sim 10^{-2}$  must be satisfied.

In this paper we seek a solution of (14) of the form

$$\Psi_{1}'=\varphi(t-z/v_{0})\cos(\Omega t-\lambda z), \quad \varphi=\varphi', \quad (15)$$

where  $v_0$  is the velocity of the nonlinear wave,

 $\omega_q \gg \Omega$ ,  $Q \gg \lambda$ .

Under these conditions we can get an explicit expression for the soliton when  $\Psi'_1 \sim 10^{-1}$ , which is less restrictive than in Refs. 11. Indeed, inserting (15) into (14), separating real and imaginary parts, and equating terms of the same order, we obtain the relation

$$\lambda = (\Omega/c) \left( 2 - c/v_0 \right),$$

between  $\Omega$  and  $\lambda$ , and the nonlinear equation

$$(d\varphi/d\tau)^2 = T_1^2 \varphi^2 - b_2^2 \varphi^4, \tag{16}$$

where

$$T_{1}^{-2} = \frac{\alpha_{1}^{2}}{c/v_{0}-1} - \Omega^{2}, \qquad b_{2}^{2} = \frac{\alpha_{1}^{2}}{4(c/v_{0}-1)} \frac{\xi_{2}}{\xi_{1}}$$

where  $T_1$  is the duration of the pulse. The contribution from the small higher-order terms can be found without difficulty by the standard procedure.

Equation (16) admits the soliton solution<sup>10</sup>

$$\varphi(\tau) = (1/b_2T_1) ch^{-1}(\tau/T_1)$$

where the soliton velocity  $v_0$  satisfies  $c > v_0 > c/2$ . The area of the pulse envelope is then given by

$$\Psi_{1}' = (1/b_{2}T_{1}) \operatorname{ch}^{-1}(\tau/T_{1}) \cos(\Omega t - \lambda z), \qquad (17)$$

which corresponds to a pulsating soliton (breather).

For frequencies  $\Omega \sim 10^8$  Hz ( $\Omega < \alpha_1$ ), all of the above conditions are satisfied for typical Rayleigh waves and low paramagnetic impurity concentrations ( $\omega_Q \sim 10^{10}$  Hz, transverse relaxation time  $T_2 \sim 10^{-5}$  s, pulse length  $T_1 \sim 10^{-6}$  s,  $c \sim 10^5$  cm/s,  $\alpha_1 \sim 10^8$  Hz, cf. Refs. 3 and 4). This suggests that the solitons may be observable experimentally.

We note that for the above parameter values the soliton length is  $\sim 1$  cm, and for given parameter values a Rayleigh wave breather moves much faster than the McCall-Hahn  $2\pi$ -soliton considered in Sec. 3.

### **5. CONCLUSIONS**

We have thus found that Rayleigh wave solitons can form during self-induced transparency. A McCall-Hahn  $2\pi$ soliton forms if the resonant particles are confined to a surface monolayer, while breathers form if the paramagnetic impurities are uniformly distributed throughout the interior of the medium. Equations (12) and (17) give the explicit form of the nonlinear waves at x = 0; for x < 0 the wave amplitudes decay exponentially at the rates  $\varkappa_l$  and  $\varkappa_l$ .

In this paper we have discussed in detail the case when the Rayleigh waves excite forbidden transitions in the impurities. Our results extend easily to the case when the Rayleigh waves excite allowed transitions in paramagnetic impurities with an effective spin S = 1. For internal plane waves, this case was studied theoretically and experimentally by Shiren for MgO crystals activated with  $Fe^{2+}$  and  $Ni^{2+}$  ions (Ref. 3), and in Ref. 4 for LiNbO<sub>3</sub>: $Fe^{2+}$  crystals. We note that MgO and LiNbO<sub>3</sub> crystals are both widely used as substrates in experiments on surface acoustic waves (see, e.g., Ref. 5).

Although our analysis of the basic properties of SIT for Rayleigh waves was carried out for an isotropic elastic halfspace, the results hold quite generally. In particular, they remain qualitatively valid for self-induced transparency of Rayleigh waves in anisotropic materials. Inverted (amplifier) media correspond to a different initial condition and can be treated analogously. This case may prove to be of interest in strongly damping media.

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