Radiative decay of collective excitations in an array of quantum dots

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(Received 22 May 1996; revised manuscript received 24 June 1996)

The problem of electromagnetic response and collective excitations in an array of quantum dots is solved in the self-consistent-field approximation using the complete system of Maxwell equations. We calculate the radiative decay of collective excitations in the system and show that it can be of the order of or larger than the damping due to collisions. We show that in previous theoretical work the effect of radiation of electromagnetic waves by the dipole collective modes of the system was underestimated by several orders of magnitude. Our results remove the discrepancy between the observed and calculated linewidths of collective modes in arrays of dots. The results are discussed in connection with the related problem of the optical properties of small polarizable particles. [S0163-1829(96)03839-8]

Electrodynamic effects due to the finite velocity of light c are normally neglected in calculations of the far-infrared (FIR) response of low-dimensional electron systems such as periodic lattices of quantum wires, dots, antidots, etc. As far as the *frequency* of plasma modes is concerned, this is certainly justified as retardation effects are determined by the parameter $(a/\lambda)^2$, which is several orders of magnitude smaller than unity in real structures [here a is the lattice period (typically smaller than 1 μm) and λ is the wavelength of light (typically 50–200 μm)]. However, the radiation effects can result in a radiative decay (Γ) of plasma modes and hence influence the *linewidth* of observed resonances. In an infinite two-dimensional electron system (2DES), the FIR radiation is coupled with the two-dimensional (2D) plasmons via a periodic grating imposed on the system,[1] and the radiative decay of 2D plasmons essentially depends on the grating coupler efficiency. As shown in Ref. 2, in real 2DES the radiative linewidth Γ is much smaller than the collisional damping γ of 2D plasmons ($\Gamma/\gamma\sim 10^{-2}$). The influence of the radiative decay on the linewidth of the single-particle cyclotron resonance (CR) in the infinite 2DES is determined[3] by the dimensionless parameter $4\pi\sigma_0/c$ (σ_0/Y_0 in SI units), where σ_0 is a static conductivity of the 2DES (Y_0 is the wave admittance of free space). In earlier papers on the CR,[4] the radiative contribution to the full linewidth was usually neglected due to a small electron mobility μ.

In the structures with the dimensionality below 2 (dots, wires, etc.) plasmons can couple directly to the radiation field. In addition, the electron mobility in modern GaAs/$\mathrm{Al}_x\mathrm{Ga}_{1-x}$As heterostructures can be very high. It would therefore be reasonable to expect a relatively large contribution of the radiation effects to the full linewidth of collective modes in these structures. However, Leavitt and Little,[5] analyzing experimental data of paper[6] on the FIR response of an array of 2D electron-gas disks in a GaAs/$\mathrm{Al}_x\mathrm{Ga}_{1-x}$As heterojunction, found (in zero magnetic field $B=0$) a negligible contribution of the radiative decay ($\Gamma/\gamma<10^{-2}$). To the best of our knowledge, after that, the problem of the radiative decay of collective modes in such structures was no longer discussed in the literature.

To solve the problem, Leavitt and Little[5] considered *a single disk* in the oblate spheroid model at $B=0$. In this model one can obtain the following expression for the radiative decay of plasmons:

$$\Gamma_0=\frac{8\epsilon^{3/2}R^3\omega_0^4}{9\pi c^3}=\frac{\pi^3 n_s^2 e^4 R}{2m^2c^3\epsilon^{1/2}}, \tag{1}$$

where e and m are the charge and the effective mass of electrons, n_s is an average area density of electrons in the disk, R is the disk radius, ϵ is the dielectric constant of a surrounding medium, and ω_0 is the frequency of the dipole plasma mode at $B=0$,

$$\omega_0^2=3\pi^2 n_s e^2/4m\epsilon R. \tag{2}$$

[The formula reported in Ref. 5 differs from Eq. (1) by an additional factor ϵ in the nominator. The correct expression should obviously have the form (1), as the dielectric constant ϵ must appear in the combinations e^2/ϵ and c^2/ϵ.] The result (1) can be understood from a simple physical consideration. An external ac electric field induces an oscillating dipole moment $d\propto\exp(-i\omega_0 t)$ in the dot. The intensity of the dipole radiation is given[7] (in vacuum) by the formula $I=4\omega_0^4d^2/3c^3$. Estimating the dipole moment as $d\sim qR$, with q being the oscillating charge, and dividing I by the energy of the oscillating dipole $\sim q^2/R$, we obtain the result coinciding with (1) up to a numerical factor.

Equations (1) and (2) are valid for *a single dot*, while in real experiments one deals with *arrays of dots*. The frequency (2) is weakly influenced by interdot interaction:[8] the corrections to (2) are proportional to $(R/a)^3\ll 1$. However, this is not the case for the radiative decay of plasma modes. If the lattice period a is much smaller than the wavelength of light $a/\lambda\ll 1$, all dipoles in the area $\sim\lambda\times\lambda$ radiate in phase, which results in a considerable increase of the total radiated power and hence the radiative decay.

In this paper we calculate the FIR response of plasma modes in a square lattice of dots in arbitrary magnetic fields, taking into account the radiative decay and show that, under real experimental conditions ($a/\lambda\ll 1$) the radiative decay of plasma modes is *several orders of magnitude larger* than that found in Ref. 5. In the structures with high electron mobility the ratio Γ/γ can be of the order of or even larger than the unity.

Let a periodic lattice of 2D disks be placed at the plane $z=0$ in a magnetic field $\mathbf{B}=(0,0,B)$, and the background dielectric constants equal to ϵ_1 at $z>0$ and ϵ_2 at $z<0$. An equilibrium electron density of 2D electrons $n_s(\mathbf{r})\delta(z)$ is a periodic function of $\mathbf{r}=(x,y)$,

$$n_s(\mathbf{r})=\sum_{k,l} n_s\vartheta(\mathbf{r}-\mathbf{a}_{k,l}), \tag{3}$$

where the sum is taken over all lattice vectors $\mathbf{a}_{k,l}=a(k,l)$ and the function ϑ describes an electron density profile inside the dots [$\vartheta(\mathbf{r})\equiv\vartheta(r)$, $\langle\vartheta(r)\rangle_{\rm dot}\equiv\langle\vartheta\rangle=1$, the angular brackets mean the average over the area of a dot, and $\vartheta(r)=0$ at $r>R$]. The electric field of an external electromagnetic wave acting on electrons, $E_\alpha^{\rm ext}\propto\exp(-i\omega t)$, $\alpha=\{x,y\}$, is assumed to be uniform and parallel to the plane $z=0$.

In the periodic lattice of dots an induced electric field is expanded in a Fourier series over reciprocal lattice vectors $\mathbf{G}_{m,n}=(2\pi/a)(m,n)$,

$$E_\alpha^{\rm ind}(\mathbf{r},z)=\sum_{\mathbf{G}} E_{\alpha,\mathbf{G}}^{\rm ind}\exp(i\mathbf{G}\cdot\mathbf{r}-\kappa_{G,j}|z|), \tag{4}$$

where $j=1$ ($j=2$) for positive (negative) z,

$$\kappa_{G,j}=(G^2-\omega^2\epsilon_j/c^2)^{1/2} \tag{5}$$

is the inverse penetration length of the induced electric field in the z direction, and $G=|\mathbf{G}|$. Using the Maxwell equations, we express $E_z^{\rm ind}$ and all components of a magnetic field $\mathbf{H}(\mathbf{r},z)$ via $E_x^{\rm ind}$ and $E_y^{\rm ind}$. Imposing the standard boundary conditions at the plane $z=0$, $E_\alpha|_{-0}^{+0}=0$, $H_x|_{-0}^{+0}=4\pi j_y/c$, and $H_y|_{-0}^{+0}=-4\pi j_x/c$, we obtain a relation between the Fourier components of the induced and total electric field and an electric current j_α at $z=0$,

$$E_{\alpha,\mathbf{G}}^{\rm ind}=\frac{4\pi i\omega}{c^2}\frac{A_{\alpha\beta}}{\det\hat{A}}j_{\beta,\mathbf{G}}, \tag{6}$$

$$j_{\alpha,\mathbf{G}}=\langle j_\alpha(\mathbf{r})e^{-i\mathbf{G}\cdot\mathbf{r}}\rangle_{\rm cell}=f\langle\sigma_{\alpha\beta}(\omega,\mathbf{r})E_\beta^{\rm tot}(\mathbf{r})e^{-i\mathbf{G}\cdot\mathbf{r}}\rangle. \tag{7}$$

Here $\sigma_{\alpha\beta}(\omega,\mathbf{r})$ is a dynamic conductivity tensor assumed to be proportional to the local electron density, $f=\pi R^2/a^2$ is the area filling factor, $\hat{A}=\hat{A}^{(1)}+\hat{A}^{(2)}$,

$$A_{\alpha\beta}^{(j)}=\frac{G_\alpha G_\beta-\delta_{\alpha\beta}\omega^2\epsilon_j/c^2}{\kappa_{G,j}}, \tag{8}$$

and $\langle\rangle_{\rm cell}$ means the average over an elementary cell.

From Eqs. (6) and (7) we obtain the integral equation

$$E_\alpha^{\rm ind}(\mathbf{r})=\frac{4\pi i\omega f}{c^2}\sum_{\mathbf{G}}\frac{A_{\alpha\beta}e^{i\mathbf{G}\cdot\mathbf{r}}}{\det\hat{A}}\langle\sigma_{\beta\gamma}(\omega,\mathbf{r}')E_\gamma^{\rm tot}(\mathbf{r}')e^{-i\mathbf{G}\cdot\mathbf{r}'}\rangle, \tag{9}$$

which relates the induced field at any point of the plane $z=0$ with the total electric field inside the dots. Assuming that the induced (and total) electric field *inside* the dots is uniform (this is an exact statement in the oblate spheroid model[5,6] and in the model of parabolically confined dots[9]), multiplying Eq. (9) by $\vartheta(r)$ and integrating over the elementary cell, we obtain the relation between the total and external electric fields inside the dots, $\zeta_{\alpha\beta}(\omega)E_\beta^{\rm tot}=E_\alpha^{\rm ext}$ where the function $\zeta_{\alpha\beta}(\omega)$ has the meaning of the response function of one dot in the lattice. In a square lattice of circular dots this relation can be written in the scalar form $E_\pm^{\rm tot}=E_\pm^{\rm ext}/\zeta_\pm(\omega)$, where $E_\pm=(E_x\mp iE_y)/\sqrt{2}$ are the field amplitudes with $\pm$ circular polarizations (the upper sign corresponds to the polarization of the CR),

$$\zeta_\pm(\omega)=1-\frac{2\pi i\omega f\langle\sigma_\pm\rangle}{c^2}\sum_{\mathbf{G}}\frac{\alpha(G)A_{\alpha\alpha}}{\det\hat{A}}, \tag{10}$$

and $\sigma_\pm=\sigma_{xx}\pm i\sigma_{xy}$. The form factor

$$\alpha(G)=|\langle\vartheta e^{i\mathbf{G}\cdot\mathbf{r}}\rangle|^2 \tag{11}$$

is determined by the Fourier components of the equilibrium electron density in the dots.

The velocity of light enters the function $\zeta_\pm(\omega)$ via the inverse field penetration lengths $\kappa_{G,j}$ given by Eq. (5). Under the condition $(a/\lambda)^2\ll1$, the terms $\omega^2\epsilon_j/c^2$ in (5) can be neglected for all nonvanishing $\mathbf{G}$. The radiative decay of plasma modes in the lattice arises from the term $\mathbf{G}=\mathbf{0}$ in Eq. (10): the factor $\kappa_{G=0,j}=-i\omega\sqrt{\epsilon_j}/c$ has vanishing real and finite imaginary parts and describes induced waves radiated from the plane $z=0$ (the sign of the imaginary part Im $\kappa_{0,j}<0$ is fixed by radiative boundary conditions at $z=\pm\infty$). The response functions $\zeta_\pm$ then assume the form

$$\zeta_\pm(\omega)=1+\frac{4\pi f\langle\sigma_\pm\rangle}{c(\epsilon_1^{1/2}+\epsilon_2^{1/2})}+\frac{2\pi if\langle\sigma_\pm\rangle}{\omega(\epsilon_1+\epsilon_2)}\sum_{\mathbf{G}\neq\mathbf{0}}G\alpha(G). \tag{12}$$

Calculating Joule's heat $Q=\langle j_\alpha(\mathbf{r})E_\alpha^{\rm tot}(\mathbf{r})\rangle_{\rm cell}=\gamma_+|E_+^{\rm ext}|^2/2+\gamma_-|E_-^{\rm ext}|^2/2$ and using the Drude expressions for the conductivity $\langle\sigma_\pm\rangle=in_se^2/[m(\omega\mp\omega_c+i\gamma)]$, we find the absorption coefficients $\gamma_\pm(\omega)$ of the structure,

$$\gamma_\pm(\omega)=f\frac{\mathrm{Re}\,\langle\sigma_\pm(\omega)\rangle}{|\zeta_\pm(\omega)|^2}=\frac{n_sfe^2\gamma}{m}\frac{\omega^2}{|\omega\{\omega\mp\omega_c+i(\gamma+\Gamma)\}-\Omega_0^2|^2}, \tag{13}$$

where ω_c is the cyclotron frequency, γ is the collision relaxation rate,

$$\Gamma=\frac{4\pi n_sfe^2}{mc(\epsilon_1^{1/2}+\epsilon_2^{1/2})},\quad a\ll\lambda, \tag{14}$$

is the radiative decay of collective excitations in *an array of dots*, and Ω_0 is the excitation frequency at $B=0$,

$$\Omega_0^2=\frac{2\pi n_sfe^2}{m(\epsilon_1+\epsilon_2)}\sum_{\mathbf{G}\neq\mathbf{0}}G\alpha(G). \tag{15}$$

The changes in transmission $\Delta T_\pm$ and reflection $\Delta R_\pm$ coefficients are proportional to $\gamma_\pm/c$.

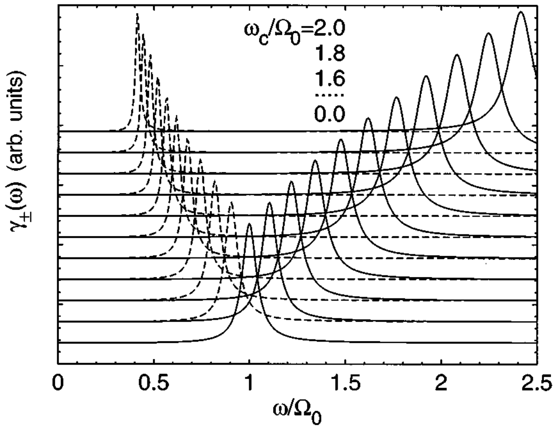

FIG. 1. Absorption coefficients $\gamma_+(\omega)$ (solid curves) and $\gamma_-(\omega)$ (dashed curves), Eq. (13), of the square lattice of dots at different magnetic fields, $\Omega_0/\gamma=20$ and $\Gamma/\gamma=0.8$.

Equation (13) describes the absorption spectrum of the square lattice of dots (Fig. 1) with the well-known[6,10] two-mode excitation spectrum with the resonance frequencies

$$\mathrm{Re}\,\omega_\pm=[(\omega_c/2)^2+\Omega_0^2]^{1/2}\pm\omega_c/2 \tag{16}$$

(for + and − circular polarizations) and the linewidths

$$\mathrm{Im}\,\omega_\pm=-\frac{\gamma+\Gamma}{2}\left\{1\pm\frac{\omega_c/2}{[(\omega_c/2)^2+\Omega_0^2]^{1/2}}\right\} \tag{17}$$

determined by the sum of the radiative decay Γ and the collisional damping γ. The ratio Γ/γ can be written as

$$\Gamma/\gamma=4\pi\sigma_0/c(\epsilon_1^{1/2}+\epsilon_2^{1/2}), \tag{18}$$

where σ_0 is the static conductivity of a system with the average electron density n_sf. At $f=0.5$, $n_s=3\times10^{11}$ cm^{-2}, $\epsilon_1=12.8$, and $\epsilon_2=1$, it exceeds unity when $\mu>5\times10^5$ cm^2/Vs.

Equation (14) correctly describes the radiative linewidth in the limit $f\rightarrow1$ of the homogeneous 2DES.[3] To obtain the special case of the single dot ($a\gg\lambda$), we apply the transformation

$$\sum_{\mathbf{G}}F(\mathbf{G})=\int\frac{a^2d\mathbf{q}}{(2\pi)^2}F(\mathbf{q})\sum_{k,l}e^{i\mathbf{q}\cdot\mathbf{a}_{k,l}} \tag{19}$$

to Eq. (10) and then take the limit $a\rightarrow\infty$. Assuming for simplicity that $B=0$, $\epsilon_1=\epsilon_2=\epsilon$, and the density profile in the dot is given by the formula

$$\vartheta(r)=\tfrac{3}{2}(1-r^2/R^2)^{1/2} \tag{20}$$

(the oblate spheroid model[5]), we obtain, at $a\gg\lambda$,

$$\zeta(\omega)=1-\frac{\omega_0^2}{\omega(\omega+i\gamma)}+\frac{i\Gamma_0\omega^2}{\omega_0^2(\omega+i\gamma)}, \tag{21}$$

with Γ_0 from Eq. (1). If $(\gamma+\Gamma_0)\ll\omega_0$, the complex zero of the function (21) describes the resonance at the frequency ω_0 with the linewidth $\gamma+\Gamma_0$. Equation (1) obtained in Ref. 5 is thus valid at $a\gg\lambda$.

Let us compare our results with experimental data of Ref. 6. The resonance linewidth observed in Ref. 6 at $B=0$ was equal to 50 GHz. The scattering linewidth was estimated in Refs. 6 and 5 to be equal to $\gamma/2\pi=16.8$ GHz (the mobility $\mu=2.5\times10^5$ cm^2/Vs). Leavitt and Little[5] estimated also the edge-scattering contribution to the resonance linewidth at $B=0$. Using the simple classical picture in which all electrons move with the Fermi velocity v_F, they obtained the mean free path for edge collisions $s=16R/3\pi$ and the mean time between edge collisions $\gamma_{\rm EC}^{-1}=s/v_F$. For the parameters of Ref. 6 ($R=1.5$ μm and $n_s=5.5\times10^{11}$ cm^{-2}) this gives $\gamma_{\rm EC}/2\pi=20.2$ GHz. The total *collisional* linewidth (momentum-relaxation plus edge-collision contributions) was thus found to be 37 GHz. The radiative decay estimated in Ref. 5 using formula (1) (multiplied by the additional factor ϵ) was found to be negligible (0.16 GHz). Calculating now the radiative contribution from Eq. (14) (we use[6] $a=4$ μm, $\epsilon_1=12.8$, and $\epsilon_2=1$), we obtain $\Gamma/2\pi=13.4$ GHz. The total linewidth turns out to be equal to 50.4 GHz, in ideal agreement with the measured value.

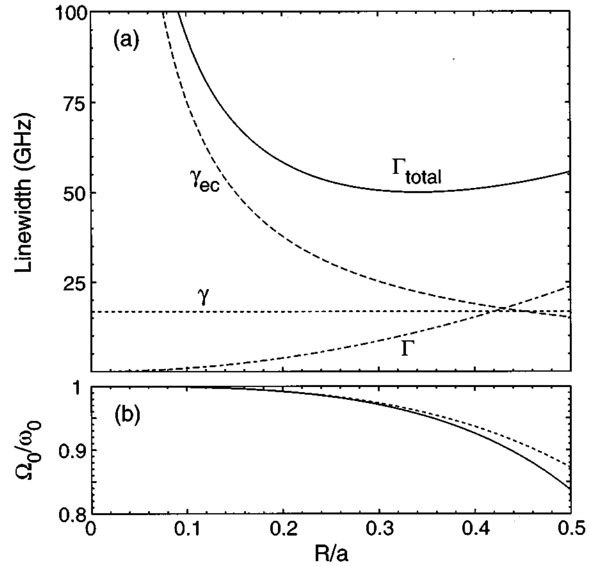

FIG. 2. (a) Linewidth and (b) frequency (23) of the plasma resonance in the square dot lattice at $B=0$ as a function of R/a; the dotted curve in (b) shows the dependence (23), taking into account only the dipole [proportional to $(R/a)^3$] correction.

Thus our results remove a discrepancy between the measured and calculated resonance linewidths and allow us to draw the conclusion that the total linewidth $\Gamma_{\rm total}=\gamma+\gamma_{\rm EC}+\Gamma$ contains three contributions: the damping due to the bulk and edge scattering and the radiative decay. Figure 2(a) demonstrates the dependence of different contributions and $\Gamma_{\rm total}$ on R/a for parameters of Ref. 6. The total linewidth has a minimum at

$$\frac{R}{a}=\frac{1}{4}\left(\frac{3c}{v_F}\frac{a_B^\star}{a}\frac{\epsilon_1^{1/2}+\epsilon_2^{1/2}}{\epsilon}\right)^{1/3}, \tag{22}$$

where $a_B^\star=\hbar^2\epsilon/me^2$ is the effective Bohr radius and $\epsilon=(\epsilon_1+\epsilon_2)/2$. Note that the radiative contribution quickly decreases with increasing a at fixed R and n_s.

Our derivation automatically takes into account the influence of the interdot interaction on the resonance frequency (15). Making use of the transformation (19), one can obtain the regular expansion of the frequency Ω_0 over the powers of R/a. For a square lattice of the dots with the profile (20) the first terms of this expansion have the form [see Fig. 2(b)]

$$\frac{\Omega_0^2}{\omega_0^2}=1-\frac{2\,\eta(3/2)}{3\pi}\left(\frac{R}{a}\right)^3-\frac{6\,\eta(5/2)}{5\pi}\left(\frac{R}{a}\right)^5+\cdots, \quad (23)$$

where $\eta(z)=\Sigma(k^2+l^2)^{-z}$ (the sum is taken over all k,l excluding $k=l=0$); $\eta(3/2)=9.03$ and $\eta(5/2)=5.09$. The dipole term proportional to $(R/a)^3$ in (23) coincides with that obtained in Ref. 8. Taking into account the interdot interaction improves the agreement between the resonance frequency at $B=0$ measured in Ref. 6 (575 GHz) and the one calculated with the help of Eq. (2) in Ref. 5 (614 GHz). Using the parameters of Ref. 6 ($R/a=0.375$), we obtain $\Omega_0/\omega_0=0.94$ and $\Omega_0=577$ GHz.

The emission of light by plasma excitations of low-dimensional electron systems can be of interest in connection with the design of light-emitting tunable FIR sources. FIR emission spectroscopy of hot 2D plasmons has been studied since about 1980 (for a recent reference see, e.g., Ref. 11). The radiative decay of 2D plasmons depends on the grating coupler efficiency, and is, however, too small.[2] As we see here, the radiative decay of magnetoplasma modes in dot lattices can be a strong effect (similar results can be obtained for wire and antidot lattices).

Formula (14) can be rewritten as ($\epsilon_1=\epsilon_2=\epsilon$)

$$\Gamma/\Gamma_0=3N/4\pi, \quad (24)$$

where N is the number of dots in the coherence area $\lambda\times\lambda$ ($N=\lambda^2/a^2$ and $\lambda=2\pi c/\omega_0\sqrt{\epsilon}$). In this form Γ does not contain the lattice period or the dot radius. This means that our assumptions on the periodicity of the spatial distribution of the dots and on their form (circular disks) are not essential for the final result. In the form (24) it should also be valid for randomly distributed small particles of an arbitrary form.

A problem similar to that considered here arises in the optics of small polarizable particles and related phenomena (surface-enhanced Raman scattering, optical properties of island metal films, etc.).[12,13] A discrepancy between the measured and predicted intensities of radiation also exists in these fields. The radiative decay of plasmons in a small metal particle as a possible way for explaining this discrepancy was discussed in the literature.[12–14] It was, however, also calculated[14] in the model of a single particle. The results obtained here show that the radiative effects can play a more important role in these phenomena, than was found in Ref. 14.

In a number of papers devoted to the theory of optical properties of adsorbate molecular layers (see, e.g., Refs. 15–17) the authors considered an interaction of light with small polarizable particles periodically arranged on a plane. In these papers, the changes in the transmission ΔT and reflection ΔR coefficients were calculated to the lowest order in $1/c$, so that the $1/c$ corrections were ignored in both the frequency and the linewidth of resonances. This does not result in a mistake in the resonance frequency, as an influence of retardation effects on the frequency is determined by the small parameter $a/\lambda\ll 1$. This can, however, result in a large mistake in the resonance linewidth, as the $1/c$ correction to the linewidth is determined by the other parameter Γ/γ, Eq. (18), which can be larger than the unity.

In conclusion, we have shown that the radiative decay of collective excitations in the dot lattices and in arrays of small polarizable particles is a much stronger effect than was assumed so far.

I thank Professor U. Rössler for reading the manuscript and useful comments. The work was supported by the Alexander von Humboldt Foundation.

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