Optical bistability in the scattering and absorption of light from nonlinear microparticles

K. M. Leung
Department of Physics, Polytechnic Institute of New York, Brooklyn, New York 11201
(Received 19 August 1985)

Intrinsic optical bistability in the scattering and absorption of light from Rayleigh-particle-sized microparticles having an intensity-dependent refractive index is theoretically investigated. For particles near plasmon resonances optical switching also occurs by sweeping the frequency across the resonance at fixed incident intensity above a certain threshold. Several orders of magnitudes of reduction in the switching intensities are possible near sharp resonances, and an optical transistor mode with a very sizable differential gain can be achieved.

Because of its potential applications as an optical memory element, an optical transistor, and logical devices for high-speed all-optical information processing, optical bistability (OB) has generated a great deal of interest.[1] In particular, intrinsic OB devices, because no resonators or external feedback structures are involved, are capable of extremely fast operations whose speeds are limited primarily only by the intrinsic response time of the nonlinear material and the transit time through the device. Materials, such as polydiacetylene P.T.S., with fast response time in the subpicosecond region and comparatively large nonlinear refractive index already exist. A decrease in the transit time can be accomplished by reducing the size of the OB element.

Our present study shows that submicron size particles can exhibit OB in the scattering and absorption of light. We consider spherical particles whose radius is small compared with the wavelength of the incident light (Rayleigh particles), and whose refractive index depends on the local electric field intensity. Near the plasmon resonance of a particle, optical switching can also occur as the frequency is swept across the resonance at fixed incident intensity above a certain threshold. If the resonance is very sharp one can find OB at intensities several orders of magnitudes lower than for the nonresonance case, and a differential gain or optical transistor mode with a gain of about 1000 can also be achieved.

Consider a dielectric sphere embedded inside a transparent host medium which has a linear dielectric constant ϵ_h. For Rayleigh particles the electrostatic approximation suffices. The electric field $\mathbf{E}_h$ in the host medium far away from the particle is supposed to be uniform. The scalar potential ϕ for $r > a$ must obey Laplace's equation and must also give an electric field equal to $\mathbf{E}_h$ when $r \rightarrow \infty$. Therefore ϕ must have the form

$$\phi(\mathbf{r}) = -\mathbf{E}_h \cdot \mathbf{r} + A \frac{\mathbf{E}_h \cdot \mathbf{r}}{r^3}, \quad r > a \ . \tag{1}$$

Inside the particle the dielectric function depends on the local intensity, i.e.,

$$\epsilon_s = \epsilon_s(\,|\,\mathbf{E}_s\,|^2)\ , \tag{2}$$

where $\mathbf{E}_s$ is the electric field within the sphere. Our basic assumption is that $\mathbf{E}_s$ is uniform throughout the particle even in the nonlinear case, and thus ϵ_s is a constant, whose value will be determined self-consistently later. Therefore the scalar potential inside also obeys Laplace's equation, and must have the form[2]

$$\phi = -B\mathbf{E}_h \cdot \mathbf{r}\ . \tag{3}$$

In Eqs. (1) and (3) we expect that the constants A and B are functions of the intensity, otherwise the calculation is exactly the same as for the linear case.[3]

Continuity of ϕ and the normal component of $\mathbf{D}$ across the surface of the sphere determines A and B

$$A = \frac{\epsilon_s/\epsilon_h - 1}{\epsilon_s/\epsilon_h + 2} a^3 \quad \text{and} \quad B = \frac{3}{\epsilon_s/\epsilon_h + 2}\ . \tag{4}$$

$\mathbf{E}_s$ is then given by

$$\mathbf{E}_s = \frac{3\mathbf{E}_h}{\epsilon_s/\epsilon_h + 2}\ . \tag{5}$$

The intensity-dependent polarizability can then be written as

$$\alpha_s = \frac{3(\epsilon_s/\epsilon_h - 1)}{4\pi(\epsilon_s/\epsilon_h + 2)} = \frac{(\epsilon_s/\epsilon_h - 1)}{4\pi} \frac{|\,\mathbf{E}_s\,|}{|\,\mathbf{E}_h\,|}\ . \tag{6}$$

The differential scattering cross section and the absorption cross section are given, respectively, by

$$\frac{d\sigma_{\rm sc}}{d\Omega} = \left[\frac{\omega}{c}\right]^4 V^2 \,|\,\alpha_s\,|^2 \sin^2\theta \tag{7}$$

and

$$\sigma_{\rm ab} = \frac{4\pi\omega V}{c} \operatorname{Im}\alpha\ ,$$

where V is the volume of the particle and θ is the angle between the scattering direction and the direction $\mathbf{E}_h$ of the polarization of the incident wave. All the formulas look exactly as in the corresponding linear case except that $d\sigma/d\Omega$ is no longer intensity independent, and $\mathbf{E}_s$ is no longer proportional to $\mathbf{E}_h$ because ϵ_s now depends on

$|\mathbf{E}_s|^2$, whose value must be determined self-consistently from Eq. (5).

To analyze the self-consistency condition we shall now specialize to the case where

$$\frac{\epsilon_s}{\epsilon_h}=\epsilon_0+\alpha\,|\mathbf{E}_s|^2 . \tag{8}$$

The effects of damping will be incorporated by considering a complex $\epsilon_0(=\epsilon_0'+i\epsilon_0'')$, while α is taken to be real. Equation (5) then gives

$$\alpha E_s^2=\frac{9\alpha E_h 2}{(p+\alpha E_s^2)^2+\epsilon_0'^2} , \tag{9}$$

where $p\equiv\epsilon_0'+2$. From Eq. (9) it is clear that optical bistability is possible only if p and α have opposite signs. Defining $x\equiv-\alpha E_s^2/p, y\equiv-9\alpha E_h^2/p^3$ and $\gamma\equiv\epsilon_0'/p$, Eq. (9) reduces to

$$y=x[(1-x)^2+\gamma^2] . \tag{10}$$

The places where jumps in x occur are given by the condition $dy/dx=0$, which gives

$$\left.\begin{matrix} x_2 \\ x_1 \end{matrix}\right\}=\tfrac{2}{3}\pm\tfrac{1}{3}(1-3\gamma^2)^{1/2}\cong\begin{cases}1-\gamma^2/2\\ \tfrac{1}{3}+\gamma^2/2 ,\end{cases} \tag{11}$$

for $\gamma^2\ll 1$. The corresponding values of y are then given by $y_2\equiv y(x_2)\cong\gamma^2$ and $y_1\equiv y(x_1)\cong\frac{4}{27}+\gamma^2/2$. The amounts of jump in x and y_1 and y_2 are roughly equal and quite sizable and are given by $1+O(\gamma^2)$. Unfortunately the critical intensity $y_c\cong\frac{4}{27}$ for the onset of OB is rather high. However, once the internal intensity is switched to its upper state where $x>1$, it remains so until y is reduced below the value $y_2\cong\gamma^2$. Therefore the holding intensity can be comparatively quite low (Fig. 1).

Next let us consider what happens at a frequency near the particle's plasmon resonance where the dielectric function can be written as

$$\epsilon_s=\epsilon_\infty\left[1-\frac{\omega_p^2}{\omega(\omega+i/\tau)}\right]+\alpha\,|\mathbf{E}_s|^2 , \tag{12}$$

where ϵ_∞ is the high-frequency dielectric constant, ω_p is the plasmon frequency, and τ is the electron relaxation time. Again we assume α to be real and frequency independent. Using Eq. (12) in Eq. (9) we find

$$\alpha E_s^2=\frac{9\alpha E_h^2\omega^4/(\epsilon_\infty+2\epsilon_h)}{\left[\omega^2-\dfrac{\epsilon_\infty\omega_p^2}{\epsilon_\infty+2\epsilon_h+\alpha E_s^2}\right]^2+\left[\dfrac{\omega}{\tau}\right]^2} . \tag{13}$$

In the linear limit the resonance frequency is given by $\omega_0=\omega_p[\epsilon_\infty/(\epsilon_\infty+2\epsilon_h)]^{1/2}$ if the resonance is sufficiently sharp. At $\omega=\omega_0$ the intensity inside the particle reaches a maximum value given by

$$(E_s^2)_{\max}=\frac{9E_h^2(\omega_0\tau)^2}{(\epsilon_\infty+2\epsilon_h)} .$$

In the nonlinear case with $\alpha>0$ Eq. (13) predicts that the plasmon frequency is lowered as the intensity inside the particle is increased. Therefore if we start at low-incident intensity with ω detuned from ω_0 toward lower frequency by an amount slightly larger than the linewidth then we expect that as the incident intensity is increased the intensity inside the particle will also increase. However, because of the nonlinearity this internal intensity will shift the effective resonance frequency toward ω and therefore allow the internal intensity to increase further. It is clear that the scattering cross section as well as the optical absorption will exhibit OB with the incident intensity as the control parameter. Optical switching and hysteresis will also occur at a fixed but sufficiently large incident intensity as the frequency is swept across ω_0.

Under most circumstances encountered experimentally it is not necessary to analyze Eq. (13) fully, as we will show below. Highly accurate results can be derived readily from a simplified form of Eq. (13).

First note that under most situations we have $|\alpha|E_s^2\ll\epsilon_\infty+2\epsilon_h$ and so the first term in the denominator of Eq. (13) can be written as

$$\omega^2-\omega_0^2+\frac{\omega_0^2\alpha E_s^2}{\epsilon_s+2\epsilon_h} . \tag{14}$$

Next we are mainly interested in frequencies close to ω_0 so that the above quantity can be approximated by

$$2\omega_0(\omega-\omega_0)+\frac{\omega_0^2\alpha E_s^2}{\epsilon_\infty+2\epsilon_h} . \tag{15}$$

Inserting this result back to Eq. (13) we find that although there are a fair number of physical parameters they can all be grouped to form three dimensionless parameters: the reduced incident intensity

$$y\equiv\frac{9\alpha E_h^2}{(\epsilon_\infty+2\epsilon_h)^2}(\omega_0\tau)^3 , \tag{16}$$

the reduced intensity inside the particle

$$x\equiv\frac{\alpha E_s^2}{(\epsilon_\infty+2\epsilon_h)}(\omega_0\tau) , \tag{17}$$

and the reduced frequency detuning

$$\delta\equiv(\omega-\omega_0)\tau . \tag{18}$$

In terms of these three parameters, we obtain from Eqs. (15) and (13) the result[4]

$$x=\frac{y}{(2\delta+x)^2+1} . \tag{19}$$

Note that x and y have the same sign as that of α and the form of Eq. (19) implies that we can confine our discussions to the self-focusing case ($\alpha>0$). The self-defocusing case can be obtained simply by changing the signs of x, y, and δ.

When the incident intensity is fixed the behavior of the internal intensity as a function of the detuning is shown in Fig. 2. To find the critical incident intensity for the onset of optical hysteresis and OB, we calculate from Eq. (19) $\partial x/\partial\delta\,|_{y=\text{const}}$ and set it equal to infinity. This gives the result

$$4\delta^2+8x\delta+3x^2+1=0 . \tag{20}$$

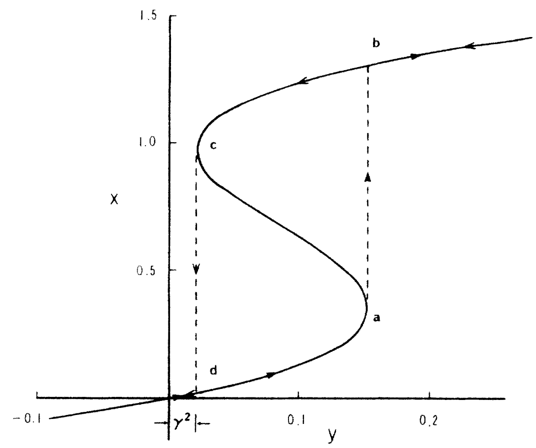

FIG. 1. Reduced electric field intensity inside a spherical Rayleigh size particle as a function of the incident intensity.

Solving x in terms of η yields

$$x=-\frac{4\delta}{3}\pm\tfrac{1}{3}(4\delta^2-3)^{1/2}\ . \tag{21}$$

At $y=y_c$ this equation must have a double root. This means that

$$\delta_c=-\frac{\sqrt{3}}{2}\ . \tag{22}$$

Putting this value in Eq. (21) implies

$$x_c=\frac{2}{\sqrt{3}}\ . \tag{23}$$

Using Eqs. (22) and (23), Eq. (19) gives the critical incident intensity

$$y_c=\frac{8\sqrt{3}}{9}\ . \tag{24}$$

In terms of the original parameters we have

$$(\alpha E_h^2)_c^r=\frac{8\sqrt{3}}{81}\frac{(\epsilon_\infty+2\epsilon_h)^{7/2}}{(\sqrt{\epsilon_\alpha}\omega_p\tau)^3}\ . \tag{25}$$

For sharp plasmon resonances where $\omega_p\tau\gg1$ the critical intensity can be several orders of magnitudes lower than for the nonresonant case discussed earlier where

$$(\alpha E_h^2)_c^{\mathrm{nr}}=\frac{4(\epsilon_0'+2)^3}{243}\ . \tag{26}$$

The curves in Fig. 2 can be easily computed by solving for δ from Eq. (19) and plotting it versus x for each fixed value of y. For $y\ll y_c$ the curve is symmetrical and Lorentzian in shape. As y increases the peak shifts toward lower frequencies and becomes asymmetrical. At $y=y_c$ the curve has a vertical tangent. For $y>y_c$ the curve bends over itself and develops a hysteresis loop.

The maximum internal intensity is exactly the same as in the linear case. The locus of points where jumps occur is given by the dotted curve. The behavior here is qualitatively similar to that of the bistable hysteresis in the cyclotron motion of a single electron.[5,6]

Next we keep the frequency fixed and study the

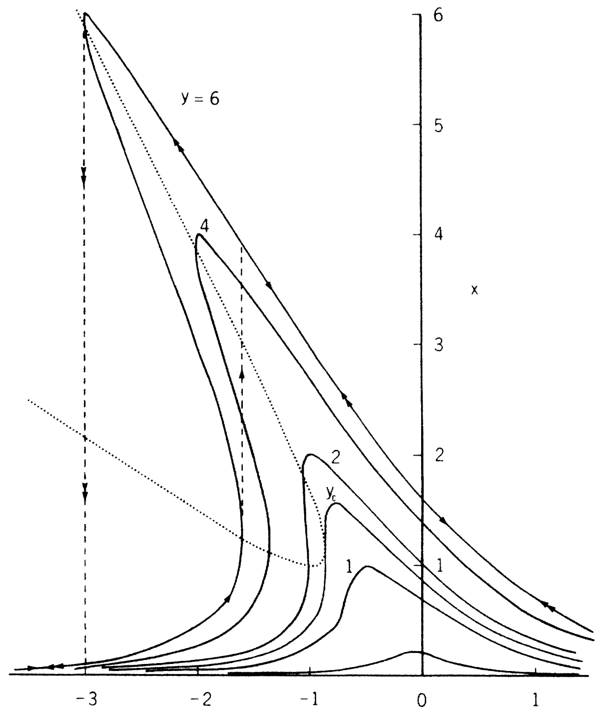

FIG. 2. Reduced internal intensity vs the detuning away from the plasmon resonance frequency for various values of the incident intensity.

behavior of the internal intensity as the incident intensity is varied. The results are shown in Fig. 3. The curves are obtained from Eq. (19) by plotting y as a function of x for different values of δ. The locus of points where jumps occur can be located for $\delta<\delta_c$ by calculating x from Eq. (21) and inserting the values into Eq. (19) for the corresponding values of y. The results are given by the dotted curve in Fig. 3. Note that the more y is above y_c, the more the mistuning can be and still obtain OB, and the more robust is the hysteresis loop. By tuning the frequency such that $\delta=\delta_c$ the hysteresis loop disappears and the curve becomes single-valued with a vertical tangent at y_c. Therefore near the critical intensity differential gain is possible. Note that the gain here can be extremely large.

To obtain a feeling for the order of magnitude of the effects discussed here we shall consider some specific examples. For the nonresonant case, Eq. (26) implies that $(\alpha E_h^2)_n^{\mathrm{nr}}$ ranges from 0.5 to about 60 for ϵ_0' equal to 2–16. Although the third-order nonlinearities in semiconductors are fairly high, unfortunately they also have a rather large dielectric constant and therefore a corresponding larger threshold for OB. Let us consider a self-defocusing material, InSb, where a value of $n_2\cong-6\times10^{-2}$ cm^{2}kW^{-1} is inferred from experiment[7] performed just below the band gap at 5 K. The threshold intensity is found to be about 100 kW/cm^2.

For the resonant case, let us consider a heavily doped n-type InSb with electron concentration 2×10^{18}/cm^3 so that the plasmon frequency lies within the range of the CO_2 laser. With $\epsilon_\infty=15.7$ and $\tau\cong8\times10^{-13}$ sec we find from Eq. (25) $(\alpha E_h^2)_c^r\cong2\times10^{-3}$. Even around the 10.6-

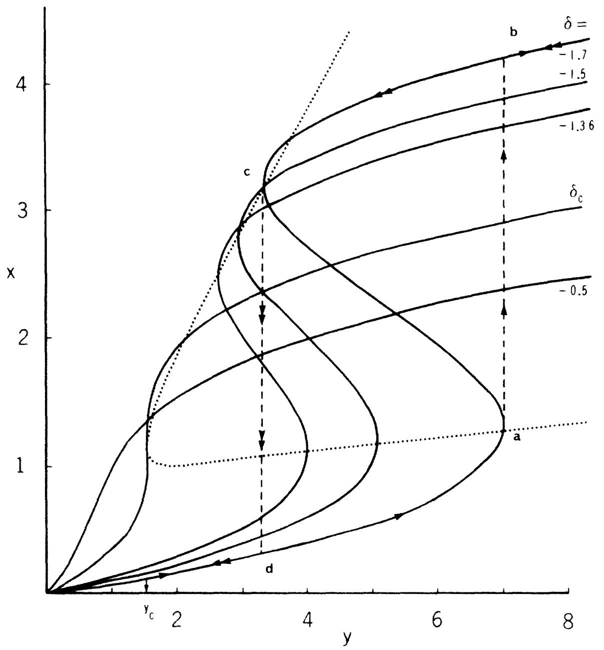

FIG. 3. Reduced internal intensity vs the reduced incident intensity for various values of the detuning. At the frequency where $\delta=\delta_c$, an optical transistor made with a very sizable differential gain can be achieved.

μm range, n_2 as high as $10^{-2}\ \mathrm{cm^2\,kW^{-1}}$ can be obtained experimentally under suitable conditions.[8] The critical intensity is found to be only about 2 kW/cm^2, and a differential gain of about 1000 can be achieved.

The idea of lowering the threshold for the onset of OB near a sharp resonance is exemplified by the recent work on the nonlinear cyclotron resonance of an electron in a Penning trap.[5,6] Although the nonlinearity, which arises from relativistic mass enhancements, is very weak for electron kinetic energy of only a few electron volts, OB can in fact be observed because of the extreme narrowness of the resonances. Resonance enhancements of electromagnetic intensities, and thus a reduction of incident power for OB in planar waveguide structures based on excitations of surface plasmons[9] and guided waves,[10] have been discussed. The latter approach has in fact been demonstrated experimentally.[11,12]

Even though our analysis here cannot be applied outside the Rayleigh particle size regime, it should be mentioned that there are very sharp "van der Hulst" resonances[13,14] for spherical particles whose sizes are large compared with the wavelength of the incident radiation. A change in the size parameter (defined as the particle circumference divided by the optical wavelength inside the particle) by a part in 10^4-10^5 can be readily detected.[15] It should be of interest to look for the kind of OB discussed here in these systems.

Note added. After submitting this article, the author noted a very recent experimental work in which a several order-of-magnitude enhancement in the optical phase-conjugated reflectivity from silver and gold colloids was reported.[16] These materials are known to have very strong resonances in the optical region. The nonlinearity involved in this experiment is basically the same as that required here for optical bistability. This is the first time that the optical Kerr coefficient has been measured for metals. Using their measured value for silver at the resonance frequency we estimate the critical intensity for switching in this system to be about 10 MW/cm^2 which should not be difficult to achieve experimentally.

This research was supported in part by the U.S. Joint Services Electronics Program under Contract No. F49620-85-C-0078. The author wishes to thank Steve Arnold for helpful discussions.

---

[1]For reviews on the subject see, for example, *Optical Bistability I*, edited by C. M. Bowden, M. Ciftan, and H. R. Robl (Plenum, New York, 1981); *Optical Bistability II*, edited by C. M. Bowden, H. M. Gibbs, and S. L. McCall (Plenum, New York, 1984); E. Abraham and S. D. Smith, Rep. Prog. Phys. **45**, 815 (1982).

[2]ϕ must also be finite at the origin.

[3]See, for example, L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon, New York, 1960), p. 42.

[4]Note that the result of Eq. (19) is formally equivalent to that of a periodically driven nonlinear oscillator, with x and y as the square of the amplitudes for the vibration and the applied force, respectively. See, for example, L. D. Landau and E. M. Lifshitz, *Mechanics*, 3rd ed. (Pergamon, New York, 1976), p. 87, but be aware of a mistake in the calculation of the critical amplitude in the earlier editions.

[5]A. E. Kaplan, Phys. Rev. Lett. **48**, 138 (1982).

[6]G. Gabrielse, H. Dehmelt, and W. Kells, Phys. Rev. Lett. **54**, 537 (1985).

[7]D. Weare, B. S. Wherrett, D. A. B. Miller, and S. D. Smith, Opt. Lett. **4**, 331 (1979).

[8]D. A. B. Miller, M. A. Mozolowski, A. Miller, and S. D. Smith, Opt. Commun. **27**, 133 (1978).

[9]G. M. Wysin, H. J. Simon, and R. J. Deck, Opt. Lett. **6**, 30 (1981).

[10]R. Reinisch and M. Neviere, Phys. Rev. B **26**, 5987 (1982); **28**, 1870 (1983).

[11]P. Martinot, A. Koster, S. Laval, and W. Carvalho, Appl. Phys. B **29**, 172 (1982).

[12]See also the review by R. Reinisch and M. Neviere, in *Nonlinear Optics: Materials and Devices*, Erice Summer School, July, 1985 (in press).

[13]H. C. van de Hulst, *Light Scattering by Small Particles* (Dover, New York, 1981).

[14]C. F. Bohren and D. R. Huffman, *Absorption and Scattering of Light by Small Particles* (Wiley, New York, 1984).

[15]S. Arnold and A. B. Pluchino, Appl. Opt. **21**, 4194 (1982).

[16]D. Picard, P. Roussignol, and C. Flytzanis, Opt. Lett. **10**, 511 (1985).