Electromagnetic energy transport via linear chains of silver nanoparticles

M. Quinten,* A. Leitner, J. R. Krenn, and F. R. Aussenegg

Institut für Experimentalphysik, Karl-Franzens Universität Graz, A-8010 Graz, Austria, and Erwin-Schrödinger Institut für Nanostrukturforschung, A-8010 Graz, Austria

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We propose the idea of a subwavelength-sized light guide represented by a linear chain of spherical metal nanoparticles in which light is transmitted by electrodynamic interparticle coupling. The light-transport properties of this system are investigated by use of model calculations based on generalized Mie theory. Considering Ag particles of 50-nm diameter, we find optimum guiding conditions for an interparticle spacing of 25 nm, and a corresponding $1/e$ signal-damping length of 900 nm is evaluated. The proposed principle of optical energy transport may be useful for subwavelength transmission lines within integrated optics circuits and for near-field optical microscopy. © 1998 Optical Society of America

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Recently a fundamental problem of integrated optics has attracted considerable interest, namely, how to transport light energy in matter structures of transverse dimensions that are considerably smaller than the corresponding wavelength of light. The main reason for the study of such subwavelength-size light-guiding principles is that the size of transmission lines within an integrated optical device is a limiting factor for further miniaturization toward a future nano-optics.

As was pointed out by Takahara *et al.*,[1] the smallest beam diameter that is possible within a given conventional dielectric optical waveguide is determined by the effective wavelength of the beam in the core material. In materials with a negative real part of the dielectric function, such as metals, there is no such cutoff dimension, as in that case the corresponding transverse wave-vector components are imaginary. Thus waveguiding systems consisting of metal structures can overcome the limits that are valid for conventional dielectric wave optics.

Takahara *et al.*[1] theoretically analyzed systems based on signal propagation along cylindrical metal structures. In this Letter we report on an alternative possibility for light guiding in subwavelength metal structures, namely, light transport via the electrodynamic interaction between a sequence of closely spaced metal nanoparticles (MNP's). The underlying principle is analyzed theoretically by model calculations for Ag spheres with respect to transmission loss as a function of particle size and interparticle spacing.

MNP's from noble metals with a small imaginary part of the dielectric function (ϵ'') are known to show strong resonances of plasma oscillation (particle plasmons) that are primarily dependent on particle shape.[2,3] Owing to these resonances, MNP's show strong light absorption; at optimum conditions the absorption cross section can exceed the mechanical cross section by nearly an order of magnitude.[4] Thus a MNP is an efficient converter of light to oscillatory energy of the electron plasma, which is a necessary condition for efficient incoupling of light into the waveguiding structure.

For the model calculations of the light-energy transport properties we assumed an infinitely long linear chain of identical Ag spheres with constant intersphere spacing. Only the first sphere in the chain is assumed to be irradiated by the light field; all other spheres get their plasma oscillation energy by coupling. As the particle line is infinitely long, it behaves as a transmission line with constant impedance. The energy flows in only one direction, and the intensity distribution along the line constantly decreases, as it is not interfered by reflections. For computational reasons, the number of spheres had to be finite, but the number was increased as long as the reflections did not contribute to the intensity along the particle chain, except at the very end (i.e., increased to 50).

We adopted the model of Gérardy and Ausloos[5] for scattering and extinction by aggregates of spherical particles of arbitrary topology and applied it to a linear chain of identical particles that were separated by a center–center distance $d_{ij} \geq 2a$ (a is the particle radius); the particles lay in the $x-y$ plane along the x axis of a reference frame (Fig. 1). A plane electromagnetic wave of wavelength λ propagating along the positive z axis is assumed to be scattered and absorbed by only the first particle. The corresponding electromagnetic fields of the incident wave, $\mathbf{E}_{\text{inc}}$ and $\mathbf{H}_{\text{inc}}$, the fields inside each particle j, $j = 1 \ldots N$, $\mathbf{E}_p(\mathbf{j})$ and $\mathbf{H}_p(\mathbf{j})$, and the fields $\mathbf{E}_{\text{sca}}(\mathbf{r}, \mathbf{j})$ and $\mathbf{H}_{\text{sca}}(\mathbf{r}, \mathbf{j})$ of the scattered wave of particle j in an arbitrary point $\mathbf{r}$ obey Maxwell's equations and can be expanded in series of vector spherical harmonics.

The scattering problem is solved when all unknown field expansion coefficients $\alpha_{nm}(j)$ and $\beta_{nm}(j)$ of the scattered waves are determined. For this purpose the electromagnetic fields $\mathbf{E}_M$ and $\mathbf{H}_M$ in the surrounding

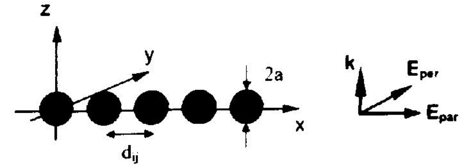

Fig. 1. Definition of the coordinate system in the calculations.

matrix are assumed to result from a linear superposition of the incident fields and the scattered fields. Then, from Maxwell's boundary conditions at the surface of sphere i, two sets of linear equations follow for the expansion coefficients $\alpha_{nm}(j)$ and $\beta_{nm}(j)$ of the scattered wave[5]:

$$\alpha_{nm}(i) - a_n(i)\sum_{j\neq i}^{N}\sum_{q=1}^{\infty}\sum_{p=-q}^{q}\alpha_{qp}(j)S_{nmqp}(i,j) + \beta_{qp}(j)T_{nmqp}(i,j) = a_n(i)\,, \quad (1)$$

$$\beta_{nm}(i) - b_n(i)\sum_{j\neq i}^{N}\sum_{q=1}^{\infty}\sum_{p=-q}^{q}\alpha_{qp}(j)T_{nmqp}(i,j) + \beta_{qp}(j)S_{nmqp}(i,j) = b_n(i)\,. \quad (2)$$

The coefficients $a_n(i)$ and $b_n(i)$ are the scattering coefficients of the single isolated sphere i for the TM and the TE modes of order n, as follows from Mie's theory.[6] The matrix elements $S_{nmqp}(i, j)$ and $T_{nmqp}(i, j)$ decrease with increasing center–center distance d_{ij} and become negligible at distances $d_{ij} \gtrsim 10a$.[7]

Applying Poynting's law, we can derive extinction and scattering cross sections as well as scattering intensities. As we are interested in the intensity distribution in the near field of the aggregate, we restrict our computations to the electromagnetic fields and intensities in the near field, according to the following procedure:

Step 1: Solve the sets of linear equations (1) and (2) for a chain of N identical spheres separated by a center–center distance d_{ij} and a plane wave propagating along the positive z axis that is polarized either parallel or normal to the chain axis (x axis) that is incident upon the first particle.

Step 2: Calculate the scattered electromagnetic fields of the particles in an arbitrary point $\mathbf{r} = (x, y, z)^T$ outside the spheres. We restrict our calculations here to $z = 0$, i.e., to the equatorial plane of the spheres.

Step 3: Coherently superpose the scattered fields of all particles $j = 1 \ldots N$ by adding $\mathbf{E}_{\mathrm{sca}}(\mathbf{r}, \mathbf{j})$ and $\mathbf{H}_{\mathrm{sca}}(\mathbf{r}, \mathbf{j})$ to $\mathbf{E}_{\mathrm{sca}}(\mathbf{r})$ and $\mathbf{H}_{\mathrm{sca}}(\mathbf{r})$.

Step 4: Calculate the intensity $I(\mathbf{r}) = \sqrt{\epsilon_0/\mu_0} \times |\mathbf{E}_{\mathrm{sca}}(\mathbf{r})|^2$, where ϵ_0 and μ_0 are the permittivity and permeability of free space, respectively.

To obtain a complete map of the intensity in the x–y plane, we vary components x and y of vector $\mathbf{r}$ continuously in a limited range. For each new set of x, y coordinates steps 2–4 are repeated.

The model calculations were performed for Ag, as this metal has the lowest optical damping properties of all metals in the visible. We used metal optical constants of Ag that were experimentally determined from measurements of MNP's.[8] As was shown in Ref. 8, for particles of 50-nm size the optical constants of the metal were different from the frequently used values of the imaginary part of the optical constant that were reported by Johnson and Christy[9]; these values were approximately a factor of 3 larger in our case, showing that the plasma oscillation in small volumes is damped more than in extended metal structures. This stronger damping is also supported by the results of recent femtosecond decay measurements.[10]

For optimum interaction with light and consequently for best incoupling of optical fields into the guide the particles should be resonant at the laser wavelength. As the resonance frequency of a nanometric metal sphere does not depend considerably on particle size (as long as the frequency is much smaller than the wavelength) for a given metal the particle resonance wavelength is practically fixed (at 380 nm for Ag). When one is choosing a layered sphere consisting of a metal shell and a dielectric core, the resonance frequency depends sensitively on the shell/core radius ratio. Choosing this ratio to be 1.515, we could model Ag spheres that were resonant at the Ar-laser line 488 nm. This resonance wavelength allows us to compare the model results with results of planned experiments. In principle, resonance wavelength tuning is also possible by use of ellipsoids with an appropriate aspect ratio instead of spheres. However, this results in significant modeling problems within the generalized Mie theory used.

Figure 2 shows a typical spatial distribution of the calculated field intensity at the beginning of a 50-sphere-long particle chain, for irradiating light polarization (a) parallel and (b) normal to the chain axis. Note the log scale in gray levels. As can be seen from the figure, at normal excitation the guiding is strongly damped, whereas at parallel excitation we observe efficient guiding. The strong polarization dependence of the guiding efficiency may result from the strong differences in near-field intensities at the position of a

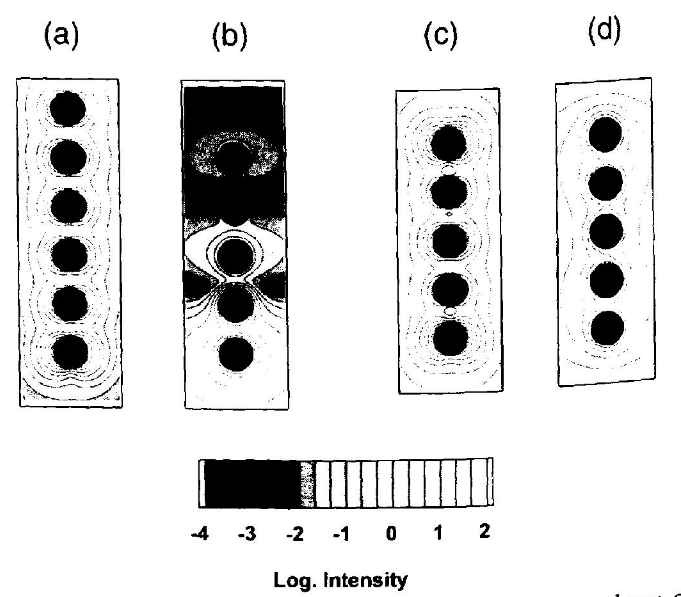

Fig. 2. x–y electric field intensity distribution along a chain of 25-nm-radius Ag nanospheres for two polarization directions: (a) parallel to the chain axis and (b) normal to the chain axis. (c), (d) Corresponding field distributions for a chain with a finite particle number of 5. Only the first particle at the bottom of the figure is assumed to be irradiated by light.

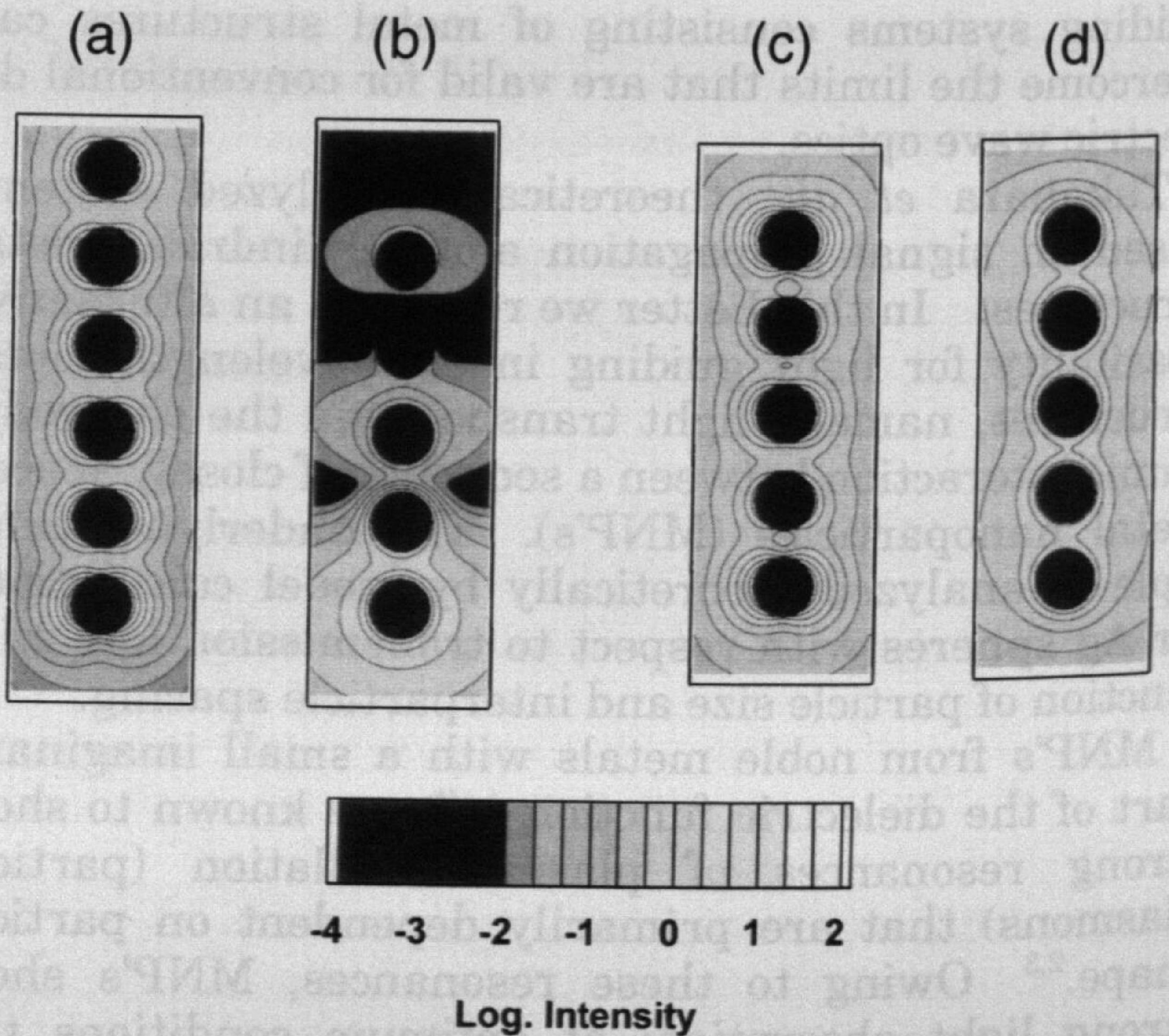

Fig. 2. $x-y$ electric field intensity distribution along a chain of 25-nm-radius Ag nanospheres for two polarization directions: (a) parallel to the chain axis and (b) normal to the chain axis. (c), (d) Corresponding field distributions for a chain with a finite particle number of 5. Only the first particle at the bottom of the figure is assumed to be irradiated by light.

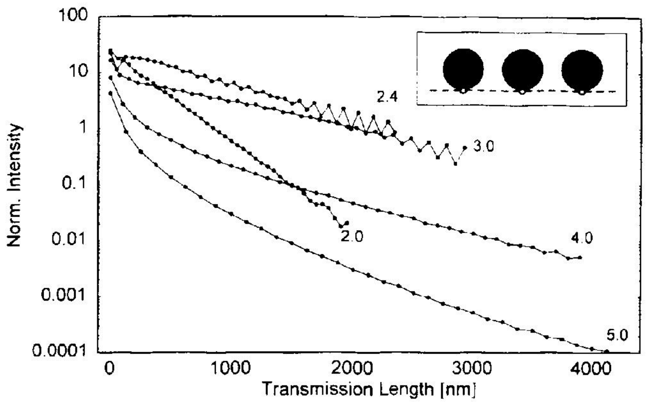

Fig. 3. Decay of field intensity in the chain axis direction. Values are taken at identical positions of each particle as shown by the open circles in the inset (corresponding to the section of Fig. 2 parallel to the axis). The given parameter d_{ij}/a is the ratio of interparticle (center–center) distance and particle radius. The intensities are normalized to the irradiated light-field intensity. Note the strong nonexponential decay for $d_{ij}/a > 3$.

Table 1. Signal-Damping Coefficient γ for the Exponential Part of the Slope per Particle and per Nanometer for Different Ratios of Particle Radius and Interparticle Distances[a]

d_{ij}/a	γ (particle^{-1})	γ (nm^{-1})	$I_{1\,\mu m}/I_{irrad}$[b]
2.0	0.19	0.0037	0.60
2.4	0.11	0.0018	6.2
3.0	0.083	0.0011	1.3
4.0			0.21
5.0			0.029

[a]The cases with $d_{ij}/a = 4.0, 5.0$ show strong nonexpontial decay; therefore a damping coefficient cannot be given. All data are for a 25-nm particle radius.
[b]Signal intensity after 1-μm guiding length, relative to the irradiated light intensity. Values >1 resonate from the resonantly enhanced field of the first particle.

neighbor particle. Figures 2(c) and 2(d) show for comparison the corresponding field distributions at a particle array with a finite particle number of 5. In this case the field intensity is distributed along the line in a complicated way, as the signal reflected at the line's end causes interference effects.

For determination of the transmission loss along a chain of metal spheres a cross section of the two-dimensional information shown in Fig. 2 was extracted. Figure 3 shows the corresponding data obtained when we cut parallel to the particle chain's axis at zero distance to each particle surface (see inset). Owing to the large number of particles in the chain (50), the signal reflected from the line's end can be neglected, as was argued above. The field intensity decays continuously with increasing distance, and some oscillations are superimposed at the beginning and at the end of the chain owing to the impedance misfit. Note that the starting values are >1 because of the resonantly enhanced field of the first particle. Transmission losses were extracted from Fig. 3 by a least-squares fit of an exponential decay applied to the cases $d_{ij}/a = 2.0$, 2.4, and 3.0, shown in Table 1. The minimum transmission loss is observed for a center–center distance/radius ratio d_{ij}/a of approximately 3. At zero interparticle distance ($d_{ij}/a = 2.0$) the transmission loss is a factor of 2.3 greater than at the optimum distance, and at larger distances the loss also increases strongly, as expected. In the optimum case $d_{ij}/a = 3.0$ for the exponential decay law $I = I_0 \exp(\gamma x)$ a signal-damping constant of $\gamma = 0.0011\,\mathrm{nm}^{-1}$ corresponding to a $1/e$ damping length of approximately 0.9 μm was evaluated. At interparticle distances greater than three times the particle radius the logarithmic signal versus distance curve deviates strongly from a straight line, indicating essential nonexponentiality, with a faster decay on the input side.

In conclusion, we have proposed the concept of subwavelength-sized light guiding by chains of metal nanoparticles by means of electrodynamic particle–particle coupling. By model calculations with the generalized Mie theory and realistic metal optical constants an optimum particle–particle distance was found, and attenuation factors were obtained in a 1-μm^{-1} range, a value of the same order of magnitude as the results obtained by other groups for cylindrical waveguide geometries.[1] It is suggested that in our case the reduced metal content should lower the losses, but the higher scattering probability should increase them. However, an exact quantitative comparison of our system with that of Ref. 1 cannot be given at the moment, owing to different data for the metal dielectric function. The obtained attenuation values are promising for the realization of short-distance signal transport by use of the principle described here.

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*Permanent address, I. Physikalisches Institut der Rheinisch–Westfälischen Technischen Hochschule Aachen, D-52056 Aachen, Germany.

References

1. J. Takahara, S. Yamagishi, H. Taki, A. Morimoto, and T. Kobayashi, Opt. Lett. **22,** 475 (1997).
2. W. Gotschy, K. Vonmetz, A. Leitner, and F. R. Aussenegg, Opt. Lett. **21,** 1099 (1996).
3. W. Gotschy, K. Vonmetz, A. Leitner, and F. R. Aussenegg, Appl. Phys. B **63,** 381 (1996).
4. C. F. Bohren and D. R. Huffman, *Absorption and Scattering Light by Small Particles* (Wiley, New York, 1983).
5. J. M. Gérardy and M. Ausloos, Phys. Rev. B **25,** 4204 (1982).
6. G. Mie, Ann. Phys. **25,** 377 (1908).
7. M. Quinten and U. Kreibig, Appl. Opt. **32,** 6173 (1993).
8. M. Quinten, Z. Phys. B **101,** 211 (1996).
9. P. B. Johnson and R. W. Christy, Phys. Rev. B **6,** 4370 (1972).
10. B. Lamprecht, A. Leitner, and F. R. Aussenegg, Appl. Phys. B **46,** 269 (1997).