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High-Frequency Waves in a Random Distribution of Metallic Nanoparticles in an External Magnetic Field

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Abstract: Propagation of magnetoplasma waves at an angle to a static magnetic field is studied for a random distribution of spherical metallic nanoparticles. A general analytical expression for dispersion relation of the system is derived and useful expressions are obtained in the limiting cases. It is found that the interaction between longitudinal and transverse modes leads to coupled modes in the vicinity of the frequency $\sqrt{f+\xi}\omega_p$, where ξ is the ratio of the volume occupied by all the nanoparticles to the entire volume, ω_p the plasma frequency of electrons inside a nanoparticle, and f a geometrical factor of order unity (1/3 for spherical nanoparticles).

Keywords: Dispersion Relation; Magnetoplasma Wave; Spherical Nanoparticle.

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1 Introduction

The interaction of electromagnetic waves with a random distribution of metallic nanoparticles is of great interest and has been studied in greater details in recent years [1–3]. In this way, Tajima et al. [4] showed that unlike an electron plasma in a metal, a random distribution of metallic nanoparticles permits propagation below the plasma cutoff of electromagnetic waves whose phase velocity is close to but below the speed of light. Parashar [5] found that a periodic lattice of nanoparticles supports an electrostatic mode of space charge oscillations with frequency lying in a narrow band and varying periodically around $\omega_p/\sqrt{3}$ with the wave number. In addition, Jain and Parashar [6] studied the dispersion characteristics of electrostatic and electromagnetic oscillations of a collection of nanoparticles in the presence of a magnetic field in the Faraday configuration (in this case, the wave vector of the incident plane wave is parallel to the magnetic field).

On the other hand, Chakhmachi and Maraghechi [7] investigated the influence of a static magnetic field in the Faraday configuration on the Raman scattering of a millimetre pump wave propagating through periodic nanoparticles. In addition, Chakhmachi [8] studied the stimulated Raman back scattering of extraordinary electromagnetic waves from the nanoparticle lattice in the presence of the static magnetic field in the Voigt configuration (in this case, the wave vector of the incident plane wave is perpendicular to the magnetic field). Furthermore, Sepehri Javan [9, 10] investigated the propagation of an intense electromagnetic waves through a periodic array of metallic nanoparticle.

As mentioned earlier, in the previous articles [6–8], much attention was devoted to the two simple geometries: Faraday configuration and Voigt configuration. The main objective of this article is a numerical calculation of the basic properties of magnetoplasma waves propagating at an angle to the magnetic field for a random distribution of spherical metallic nanoparticles, where nanoparticles are separated by distances greater than their characteristic size and concentration of metallic nanoparticles is lower than the percolation threshold. In other words, the present calculation is, in fact, correct only to first order in the volume fraction of nanoparticles. In this way, we use small values of filling factor ξ in the calculation because large volume fraction probably causes the percolation effect. Moreover, we note that characteristic size of nanoparticles is small compared to the wavelength in the effective medium. In addition, we find useful analytic expressions for propagation in some special cases such as unmagnetised, collisionless, and the Faraday and Voigt geometries. Some of the formulas are well known, and we repeat them only for the sake of completeness.

This article is organised as follows: In Section 2, we set the basic equations concerning the problem. Then, we obtain a general analytical expression for the dispersion relation of electromagnetic oscillations of the system in the presence of the external magnetic field and collisional

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effects. In Section 3, the dispersion relation of magnetoplasma waves is analysed in special cases. In Section 4, the numerical results are discussed, and finally, Section 5 contains our conclusions.

2 Formulation of Problem

Consider, for the moment, a collection of spherical metallic nanoparticles with the uniform electron density equal to all nanoparticles. We now perturb this equilibrium with a plane electromagnetic wave and study the medium response. We assume that the wave frequency is high enough that the ions can be considered as stationary. Without losing any generality in our plane electromagnetic wave solutions, we have been taking $\mathbf{B}_0$ in the z-direction, and the $\mathbf{k}$-vector to have components only in the x- and z-directions, as shown in Figure 1.

Under the influence of electric field of electromagnetic wave, the electron cloud of the nanoparticles is displaced and leads to the creation of surface charges, positive where the cloud is lacking, negative where it is concentrated. However, one has to keep in mind that all the electrons of the nanoparticles are moving collectively while under the effect of the field. Such collective oscillation leads to localised surface plasmons, in contrast to free plasmons occurring in the bulk metals [11]. Because of this dipolar charge repartition, there arises a strong restoring force due to plasma electron space charge, which is different from but has similarity with the longitudinal plasma charge restoring force. Therefore, for a plasmonic nanoparticle, the equation of motion of an electron in the $\mathbf{r}$-direction, is

$$\left(\frac{\mathrm{d}^2}{\mathrm{d}t^2}+\gamma\frac{\mathrm{d}}{\mathrm{d}t}+f\omega_p^2\right)\mathbf{r}=-\frac{e}{m}\mathbf{E}-\frac{e}{m}\mathbf{u}\times\mathbf{B}_0, \tag{1}$$

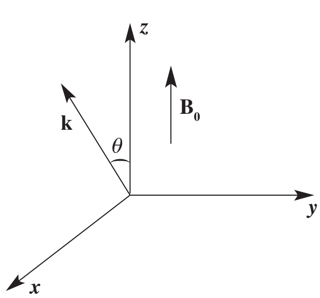

Figure 1: For wave propagation in a random distribution of magnetised nanoparticles, the angle of propagation θ with respect to the static magnetic field is important. Here, we assume that $\mathbf{B}_0$ is along the z-axis of a rectangular coordinate system and that $\mathbf{k}$ is in the x–z plane, as shown.

where $-f\omega_p^2\mathbf{r}$ is the restoring force on electron and $\sqrt{f}\omega_p$ (with $f=1/3$) the surface plasmon frequency of the spherical metallic nanoparticle. Furthermore, $\mathbf{r}=x\mathbf{e}_x+y\mathbf{e}_y+z\mathbf{e}_z$, $\mathbf{u}=u_x\mathbf{e}_x+u_y\mathbf{e}_y+u_z\mathbf{e}_z$, and $\omega_p^2=e^2n_0/\epsilon_0 m$. Moreover, e and m are electron charge and mass and γ phenomenological damping or the damping constant due to scattering of metal electrons, which may lead to wave absorption. It should be noted that the value of γ in very small particles is indeed expected to increase beyond its value in bulk samples, since the mean free path of the electrons is reduced as a result of collisions with the surfaces [12]. Resolving (1) into x, y, and z components and using the operator $\mathrm{d}/\mathrm{d}t=-i\omega$, we obtain

$$[\omega(\omega+i\gamma)-f\omega_p^2]x=\frac{e}{m}E_x-i\omega\omega_c y, \tag{2}$$

$$[\omega(\omega+i\gamma)-f\omega_p^2]y=\frac{e}{m}E_y+i\omega\omega_c x, \tag{3}$$

$$[\omega(\omega+i\gamma)-f\omega_p^2]z=\frac{e}{m}E_x, \tag{4}$$

where $\omega_c=eB_0/m$ is the electron cyclotron frequency. Using (2) and (3), we obtain

$$x=\frac{e}{m}\frac{\chi_{xx}E_x+\chi_{xy}E_y}{\Gamma}, \tag{5}$$

$$y=\frac{e}{m}\frac{\chi_{yx}E_x+\chi_{yy}E_y}{\Gamma}, \tag{6}$$

$$z=\frac{e}{m}\frac{\chi_{zz}E_z}{\Xi}, \tag{7}$$

where $\Xi=\omega(\omega+i\gamma)-f\omega_p^2$, $\Gamma=[\omega(\omega+i\gamma)-f\omega_p^2]^2-\omega^2\omega_c^2$, $\chi_{xx}=\chi_{yy}=\Xi$, $\chi_{zz}=1$, $\chi_{xy}=-\chi_{yx}=-i\omega\omega_c$. On simplifying (5)–(7), we obtain the velocity of electron cloud as

$$\mathbf{u}=-i\omega\frac{e}{m}\left[\frac{\chi_{xx}E_x+\chi_{xy}E_y}{\Gamma}\mathbf{e}_x+\frac{\chi_{yx}E_x+\chi_{yy}E_y}{\Gamma}\mathbf{e}_y+\frac{\chi_{zz}E_z}{\Xi}\mathbf{e}_z\right] \tag{8}$$

Therefore, the current density $\mathbf{J}$ can be written as

$$\mathbf{J}=i\xi\epsilon_0\omega\omega_p^2\left[\frac{\chi_{xx}E_x+\chi_{xy}E_y}{\Gamma}\mathbf{e}_x+\frac{\chi_{yx}E_x+\chi_{yy}E_y}{\Gamma}\mathbf{e}_y+\frac{\chi_{zz}E_z}{\Xi}\mathbf{e}_z\right]. \tag{9}$$

By using the ohm's law ($\mathbf{J}=\underline{\sigma}\cdot\mathbf{E}$), we find the complex frequency-dependent tensor of electrical conductivity as

$$\underline{\sigma}=\frac{i\xi\epsilon_0\omega\omega_p^2}{\Gamma}\begin{pmatrix}\chi_{xx} & \chi_{xy} & 0\\ \chi_{yx} & \chi_{yy} & 0\\ 0 & 0 & \chi_{zz}\Gamma/\Xi\end{pmatrix}, \tag{10}$$

This tensor conductivity can be substituted into the wave equation to construct a dispersion relation. The relevant equations for the electromagnetic wave propagation are Faraday's law and Ampere's law coupled with Ohm's law. These are found in the following equations:

$$\nabla\times\mathbf{E}=i\omega\mathbf{B}, \tag{11}$$

$$\nabla\times\mathbf{B}=\mu_0\mathbf{J}-i\omega\mu_0\epsilon_0\mathbf{E}, \tag{12}$$

$$\mathbf{J}=\underline{\sigma}\cdot\mathbf{E}. \tag{13}$$

Combining the above-mentioned equations leads to the wave equation

$$\nabla^2\mathbf{E}-\nabla(\nabla.\mathbf{E})+\frac{\omega^2}{c^2}\left(\boldsymbol{I}+i\frac{\underline{\sigma}}{\epsilon_0\omega}\right)\cdot\mathbf{E}=0, \tag{14}$$

where $\boldsymbol{I}$ is just the identity tensor or, in index notation, the matrix with ones along the main diagonal and zeros elsewhere. Since a phase dependence $\exp(i\mathbf{k}\cdot\mathbf{r}-i\omega t)$ is assumed, the above-mentioned equation can be written in algebraic form as

$$k^2\mathbf{E}-\mathbf{k}(\mathbf{k}.\mathbf{E})-\frac{\omega^2}{c^2}\underline{\epsilon}\cdot\mathbf{E}=0, \tag{15}$$

where we have a dielectric tensor, denoted by $\underline{\epsilon}$, given by

$$\underline{\epsilon}=\boldsymbol{I}+i\frac{\underline{\sigma}}{\epsilon_0\omega}.$$

Since we are using tensor notation, we re-express the left-hand side of (15) in tensor notation. Therefore, we find

$$\left(\frac{\omega^2}{c^2}\underline{\epsilon}-k^2\underline{\kappa}\right)\cdot\mathbf{E}=0, \tag{16}$$

where $\underline{\kappa}$ is the tensor defined by $\underline{\kappa}=\boldsymbol{I}-\mathbf{kk}/k^2$. Remembering that we have chosen $k_y=0$, so $\mathbf{k}=k\sin\theta\mathbf{e}_x+k\cos\theta\mathbf{e}_z$, where θ is the angle of the wave vector with respect to the z-axis. The dispersion relation is then derived from the requirement that the determinant of the tensor quantity in parentheses in (16) be zero. Thus, the dispersion of the system can be written as

$$\det\begin{pmatrix}S-n^2\cos^2\theta & iD & n^2\cos\theta\sin\theta\\ -iD & S-n^2 & 0\\ n^2\cos\theta\sin\theta & 0 & P-n^2\sin^2\theta\end{pmatrix}=0, \tag{17}$$

where $S=1-\xi\omega_p^2\Xi/\Gamma$, $D=i\xi\chi_{xy}\omega_p^2/\Gamma$, $P=1-\xi\omega_p^2/\Xi$, and $n=kc/\omega$. After doing some algebra, (17) becomes

$$n^2=1-\frac{\frac{\xi\omega_p^2}{\omega^2}\left(1-\frac{\xi\omega_p^2}{\Xi}\right)}{\frac{\Xi}{\omega^2}\left(1-\frac{\xi\omega_p^2}{\Xi}\right)-\frac{\omega_c^2}{2\Xi}\sin^2\theta\pm\Upsilon} \tag{18}$$

where

$$\Upsilon=\sqrt{\frac{\omega_c^4}{4\Xi^2}\sin^4\theta+\frac{\omega_c^2}{\omega^2}\left(1-\frac{\xi\omega_p^2}{\Xi}\right)^2\cos^2\theta}. \tag{19}$$

The above-mentioned equation is the original result of this work, the (complex) dispersion relation of electromagnetic waves in a nanoparticle plasma in the presence of a static magnetic field. The ± sign indicates the left- and right-hand polarisation waves, respectively. We note that structure of (18) is similar to the well-known Altar–Appleton–Hartree formula, which has been extensively applied to radio waves in the ionosphere [13, 14]. In Section 3, we study this dispersion relation in different cases.

3 Analysis of the Dispersion Relation in Different Cases

In this section, we investigate the solution of (18) for different cases as below.

Case 1. If we neglect the external magnetic field effect, (18) leads to the following dispersion equation for propagation of electromagnetic waves in an unmagnetised nanoparticle plasma

$$n^2=1-\frac{\xi\omega_p^2}{\omega(\omega+i\gamma)-f\omega_p^2}. \tag{20}$$

This result is the same as (3) derived by Tajima et al. [4].

Case 2. In this case, the wave vector $\mathbf{k}$ is parallel to the magnetic field $\mathbf{B}_0$ (Faraday configuration), i.e. we have $\theta=0$. Therefore, (18) becomes

$$n^2=1-\frac{\xi\omega_p^2}{\omega(\omega+i\gamma)-f\omega_p^2\pm\omega\omega_c}. \tag{21}$$

The above-mentioned equation has four real solutions. On the other hand, since a factor $[\omega(\omega+i\gamma)-(f+\xi)\omega_p^2]$ has cancelled out in the numerator and denominator of (18), we also have a solution

$$\omega(\omega+i\gamma)=(f+\xi)\omega_p^2. \tag{22}$$

This, of course, means that in the Faraday configuration plasmons do not interact with other types of excitations [15]. If we assume $\gamma=0$ and $\xi=4\pi\ell/3$ (where $\ell=(R/\mathrm{d})^3$, R is the radius of each nanoparticle and d the separation between nanoparticles [7]), from (21), we get the result of [4].

Case 3. Here, we want to solve (18) by considering $\theta\simeq 0$. The result gives the dispersion relation for quasi-parallel propagation. In this case, the term containing $\cos^2\theta$ dominates in (18). After doing some algebra, one obtains

$$n^2=1-\frac{\xi\omega_p^2}{\omega(\omega+i\gamma)-f\omega_p^2\pm\omega\omega_c\cos\theta}, \tag{23}$$

where for the upper and lower signs, we call these the quasi-parallel, left-hand circularly polarised and the quasi-parallel, right-hand circularly polarised, respectively.

Case 4. Now, we obtain a dispersion relation for the Voigt configuration, where the wave vector $\mathbf{k}$ is perpendicular to the magnetic field $\mathbf{B}_0$ ($\theta=\pi/2$). Therefore, (18) becomes

$$n^2=1-\frac{\frac{\xi\omega_p^2}{\omega^2}\left(1-\frac{\xi\omega_p^2}{\Xi}\right)}{\frac{\Xi}{\omega^2}\left(1-\frac{\xi\omega_p^2}{\Xi}\right)-\frac{\omega_c^2}{2\Xi}\pm\frac{\omega_c^2}{2\Xi}}, \tag{24}$$

which also has two solutions. First, we have

$$n^2=1-\frac{\xi\omega_p^2}{\omega(\omega+i\gamma)-f\omega_p^2}, \tag{25}$$

which is the same as those presented by Tajima et al. [4]. The other solution of (24) is

$$n^2=1-\frac{\frac{\xi\omega_p^2}{\omega^2}\left(1-\frac{(f+\xi)\omega_p^2}{\omega(\omega+i\gamma)}\right)}{\frac{\omega+i\gamma}{\omega}\left(1-\frac{f\omega_p^2}{\omega(\omega+i\gamma)}\right)^2\left(1-\frac{\xi\omega_p^2}{\omega(\omega+i\gamma)-f\omega_p^2}\right)-\frac{\omega_c^2}{\omega(\omega+i\gamma)}}. \tag{26}$$

If we assume $\gamma=0$, $f=0$, and $\xi=1$, from (26), we find the low-frequency and high-frequency extraordinary modes of an electron plasma in a metal. However, it is easy to find that our dispersion relation in the limiting case of the Voigt configuration, i.e. (26) does not agree with the result obtained by Chakhmachi [8]. We note the extraordinary modes of an electron plasma in a metal cannot derived from the Chakhmachi result, i.e. (10) in [8].

Case 5. Finally, we consider the case quasi-transverse dispersion ($\theta\simeq\pi/2$). Here, transverse means $\mathbf{k}$ is nearly perpendicular to the static magnetic field. For quasi-transverse propagation, the first term in Υ dominates, that is

$$\frac{\omega_c^4}{4\Xi^2}\sin^4\theta\gg\frac{\omega_c^2}{\omega^2}\left(1-\frac{\xi\omega_p^2}{\Xi}\right)^2\cos^2\theta. \tag{27}$$

In this case, a binomial expansion of Υ gives

$$\begin{aligned}\Upsilon&=\frac{\omega_c^2}{2\Xi}\sin^2\theta\left\{1+\frac{4\Xi^2}{\omega^2\omega_c^2}\left(1-\frac{\xi\omega_p^2}{\Xi}\right)^2\frac{\cos^2\theta}{\sin^4\theta}\right\}^{1/2}\\&\simeq\frac{\omega_c^2}{2\Xi}\sin^2\theta+\frac{\Xi}{\omega^2}\left(1-\frac{\xi\omega_p^2}{\Xi}\right)^2\cot^2\theta.\end{aligned} \tag{28}$$

Substitution of the above-mentioned equation into (18) shows that the generalisation of the nonmagnetic modes dispersion to angles in the vicinity of $\pi/2$ is

$$n_+^2=1-\frac{\xi\omega_p^2\sin^2\theta}{\omega(\omega+i\gamma)-f\omega_p^2-\xi\omega_p^2\cos^2\theta}. \tag{29}$$

The subscript + here means that the positive sign has been used in (18). This mode may be called the quasi-transverse-nonmagnetic modes. Choosing the − sign in (18) gives the quasi-transverse-extraordinary modes. We have

$$n_-^2=1-\frac{\frac{\xi\omega_p^2}{\omega^2}\left(1-\frac{(f+\xi)\omega_p^2}{\omega(\omega+i\gamma)}\right)}{\frac{\omega+i\gamma}{\omega}\left(1-\frac{f\omega_p^2}{\omega(\omega+i\gamma)}\right)^2\left(1-\frac{\xi\omega_p^2}{\omega(\omega+i\gamma)-f\omega_p^2}\right)-\frac{\omega_c^2}{\omega(\omega+i\gamma)}\sin^2\theta}. \tag{30}$$

4 Numerical Results and Discussion

Now, we present the simulation results of dispersion relation of magnetoplasma waves in a magnetised spherical nanoparticle plasma and investigate their dependence on the parameters θ, as shown in Figures 2 and 3 for $\xi=0.1$. We note that the most interesting feature of propagation in the intermediate geometry is the coupling of plasmons with transverse waves. While in the Faraday configuration the dispersion curves may intersect, for any finite-angle θ,

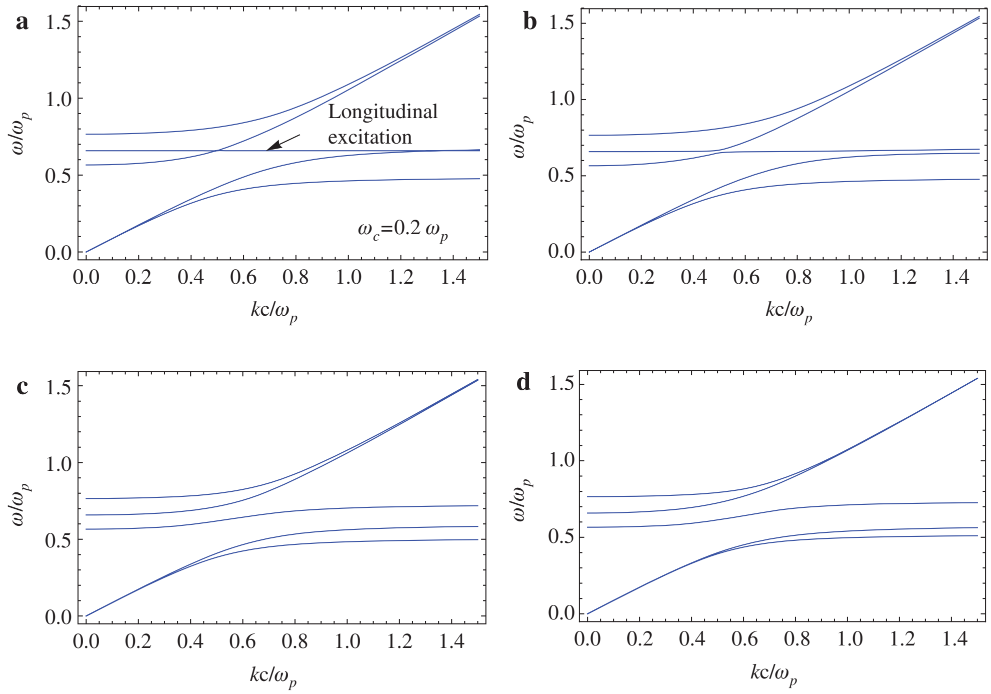

Figure 2: Dispersion curves for magnetoplasma waves propagating in a random distribution of magnetised spherical nanoparticles with $\omega_c/\omega_p=0.2$ and $\xi=0.1$, when (a) $\theta=0°$, (b) $\theta=10°$, (c) $\theta=60°$, and (d) $\theta=90°$.

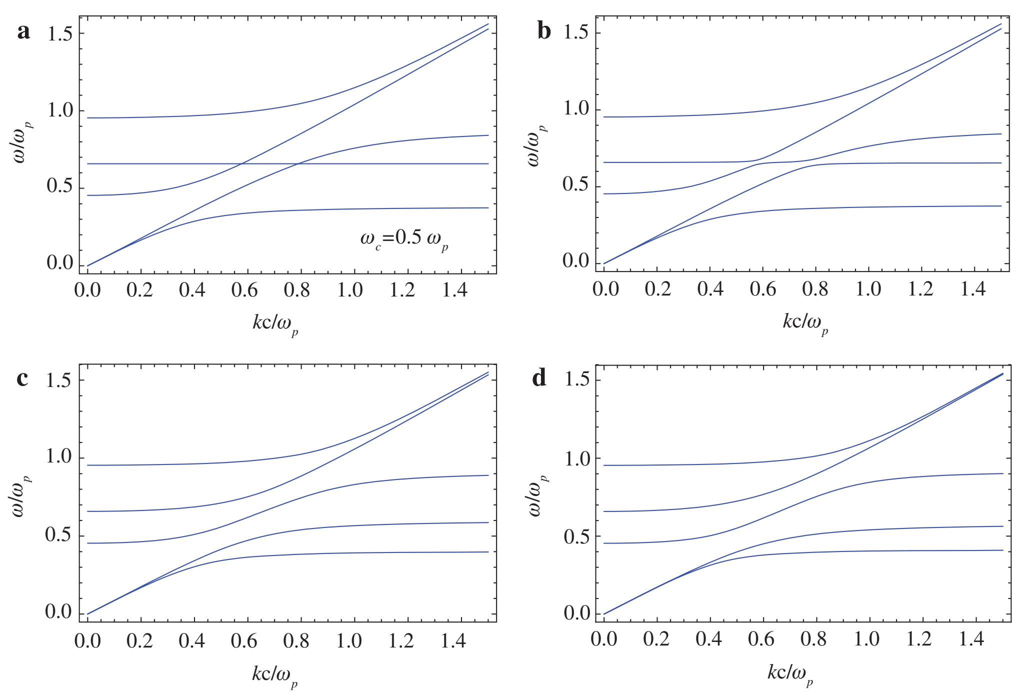

Figure 3: Same as Figure 2 but for $\omega_c/\omega_p=0.5$.

a repulsion takes place. One can see that by increasing θ, repulsion increases. It is clear that there is one intersection of modes (i.e. one coupling of longitudinal excitations and transverse magnetoplasma waves) in Figure 2(b), for $\omega_c=0.2\omega_p$, and two intersections (i.e. two coupling of plasmon mode and transverse magnetoplasma waves) in Figure 3(b), for $\omega_c=0.5\omega_p$. The reason for this behaviour becomes apparent from (22) and (23) (assuming small θ).

The point of intersection is given by the solution of these equations. For $\gamma = 0$, we obtain

$$\frac{k^2c^2}{\omega_p^2} = \frac{(f+\xi)^{3/2}\omega_c\cos\theta}{(f+\xi)^{1/2}\omega_c\cos\theta \pm \xi\omega_p}. \tag{31}$$

Thus, there is always an intersection of modes for the upper sign. For the lower sign, however, there is a real solution only if $(f+\xi)^{1/2}\omega_c \cos\theta > \xi\omega_p$.

5 Conclusion

In summary, we have studied the propagation of linear electromagnetic waves in a random distribution of spherical metallic nanoparticles, in the presence of a static magnetic field and collisional effects. We have derived a general analytical expression for the dispersion relation of the system. Considering different cases such as unmagnetised, collisionless, and Faraday configuration cases, some formulas presented in the previous studies have been obtained. However, we have found that our dispersion relation in the limiting case of the Voigt configuration does not agree with the result obtained by Chakhmachi [8]. In addition, we have presented in graphical forms the basic properties of magnetoplasma waves propagating at arbitrary angle θ to the magnetic field. We have shown that for $\theta \neq 0°$ coupling of transverse magnetoplasma waves and plasmon mode takes place.

References

[1] U. Kreibig and M. Vollmer, Optical Properties of Metal Clusters, Springer Series in Material Science, Springer, Berlin 1995, Vol. 25.
[2] V. M. Shalaev, Optical Properties of Nanostructured Random Media, Topics in Applied Physics, Springer, Berlin 2002, Vol. 82.
[3] A. Stalmashonak, G. Seifert, and A. Abdolvand, Ultra-Short Pulsed Laser Engineered Metal-Glass Nanocomposites, SpringerBriefs in Physics, Springer, New York 2013.
[4] T. Tajima, Y. Kishimoto, and M. C. Downer, Phys. Plasmas **6**, 3759 (1999).
[5] J. Parashar, Phys. Plasmas **16**, 093106 (2009).
[6] S. Jain and J. Parashar, J. Opt. **40**, 71 (2011).
[7] A. Chakhmachi and B. Maraghechi, Phys. Plasmas **18**, 022102 (2011).
[8] A. Chakhmachi, Phys. Plasmas **20**, 062104 (2013).
[9] N. Sepehri Javan, J. Appl. Phys. **118**, 073104 (2015).
[10] N. Sepehri Javan, Phys. Plasmas **22**, 093116 (2015).
[11] A. V. Zayats, I. I. Smolyaninov, and A. A. Maradudin, Phys. Rep. **408**, 131 (2005).
[12] S. A. Maier, Plasmonic: Fundamentals and Applications, Springer, New York 2007.
[13] W. P. Allis, S. J. Buchsbaum, and A. Bers, Waves in Anisotropic Plasmas, MIT Press, Cambridge 1963.
[14] D. G. Swanson, Plasma Waves, IOP, London 2003.
[15] A. Moradi, J. Appl. Phys. **107**, 066104 (2010).