

ical behavior can very likely be ascribed to the use of too high frequencies (i.e., magnetic fields) and of improper ferromagnets, where no true exchange critical regime exists.

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## Temperature Dependence of the Plasma Frequency of Two-Component Ionic Fluids

Marc Baus

*Chimie-Physique II, Université Libre de Bruxelles, B-1050 Bruxelles, Belgium*

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In two recent molecular-dynamics experiments on two-component ionic fluids it was observed that the plasma frequency was shifted with respect to its mean-field value. This effect is explained here on the basis of a microscopic theory. The shift is shown to exhibit a strong plasma parameter dependence.

Recently, the fastly growing literature on Coulomb systems has been enriched with a new molecular-dynamics (MD) study of the time-dependent fluctuations in a  $H^+ - He^{++}$  mixture.<sup>1</sup> It was observed there that the infinite-wavelength ( $k = 0$ ) plasma oscillations occur in this system at a frequency which is shifted both with respect to the mean-field prediction and with respect to the prediction from phenomenological hydrodynamics. The observed oscillation frequency was found to agree nevertheless with a sum-rule analysis predicting a plasma frequency independent of the

system's plasma parameter.

In this Letter I analyze the above experiment on the basis of a microscopic theory<sup>2</sup> which has previously been applied to the one-component plasma (OCP)<sup>3-5</sup> and was recently extended to multicomponent Coulomb systems.<sup>6</sup> I consider a system of two mobile species ( $\sigma = 1, 2$ ) of mass  $m_\sigma$ , charge  $e_\sigma$ , and average number density  $n_\sigma$  immersed in an inert neutralizing background, and characterized by the plasma parameters  $\Gamma_\sigma = (\frac{4}{3}\pi)^{1/3} e_\sigma^2 n_\sigma^{1/3} \beta$ . When  $e_1 e_2 > 0$  such a system will be called a binary ionic mixture (BIM) and

it provides a simple model, for instance, for the  $H^+ - He^{++}$  mixture.<sup>1</sup> When  $e_1 e_2 < 0$  and

$$\sum_{\sigma=1,2} e_{\sigma} n_{\sigma} = 0,$$

I will speak instead of a two-component plasma (TCP). Here no neutralizing background is required. The TCP provides a simple model for a genuine plasma or a molten salt.<sup>7</sup> Formally all

these two-component ionic fluids can be treated in the same manner. The physical behavior of these systems is, however, very different. Let us start our analysis by considering the dispersion equation for the collective modes of these two-component ionic fluids. In the present theory<sup>2,3</sup> this dispersion equation reads as follows:  $\Delta(\vec{k}, z) \equiv \det[zI - \Omega(\vec{k}, z)]$ , where  $\Omega(\vec{k}, z)$  is the transport matrix which is related to the memory function  $\Sigma(\vec{k}, z)$  by

$$\Omega_{\alpha\beta}(\vec{k}, z) = \langle \alpha | \Sigma(\vec{k}, z) | \beta \rangle + \langle \alpha | \Sigma(\vec{k}, z) Q [z - Q \Sigma(\vec{k}, z) Q]^{-1} Q \Sigma(\vec{k}, z) | \beta \rangle, \quad (1)$$

where, as usual,  $Q$  projects out the hydrodynamic states  $|\alpha\rangle$ . In the present multispecies case I will use<sup>6</sup> multifluid hydrodynamic variables,  $\alpha \equiv (\delta, \sigma)$ , corresponding to the total number, momentum, and excess kinetic energy ( $j = 1-5$ ) of each species ( $\sigma = 1, 2$ ). For isotropic fluids  $\Delta(\vec{k}, z)$  can be factorized<sup>3</sup> so as to separate the transverse shear modes from the longitudinal modes which, at  $k = 0$ , can in turn be separated into heat modes and density ( $n$ )—longitudinal momentum ( $l$ ) modes. At infinite wavelengths ( $k = 0$ ) the dispersion equation for the latter modes turns out to be very simply given by

$$\Delta_{n,l}(\vec{k} = \vec{0}, z) \equiv z^2(z^2 - \Omega_p^2) + i\nu_l(z)z(z^2 - \Omega^2) = 0. \quad (2)$$

This dispersion equation, which is *exact*, is identical to what one would get from more phenomenological considerations except that instead of having a phenomenological relaxation frequency we have here a microscopically defined, frequency-dependent relaxation frequency  $\nu_l(z)$ . This quantity is defined by

$$\nu_l(z) = i \sum_{\sigma=1,2} \Omega_{ll}^{\sigma\sigma}(\vec{k} = \vec{0}, z), \quad (3)$$

where  $\Omega_{ll}^{\sigma\sigma}$  is the element of the transport matrix of Eq. (1) corresponding to  $\alpha = \beta = (l, \sigma)$ ,  $l$  denoting the longitudinal momentum state. All the microscopic information necessary for describing the interspecies longitudinal momentum relaxation has been lumped together into the single quantity  $\nu_l(z)$ . Contrary to what happens in a phenomenological treatment  $\nu_l(z)$  is, however, not a positive parameter but in general a complex function of  $z$ . From the symmetry properties of the transport matrix one can show that at real frequency  $\omega$ ,  $z = \omega + i0$ , the real ( $\nu_R$ ) and imaginary ( $\nu_I$ ) parts of  $\nu_l$  are, respectively, even and odd functions of  $\omega$ :

$$\nu_l(\pm\omega) = \nu_R(\omega) \pm i\nu_I(\omega). \quad (4)$$

For stability reasons  $\nu_R(\omega)$  has moreover to be positive. Now, if the charge relaxation modes were genuine hydrodynamical modes a knowledge of  $\nu_l(\omega = 0) \equiv \nu_R(0) > 0$  would suffice and the phenomenological approach would be qualitatively correct. The high-frequency plasma oscillations require instead, even at  $k = 0$ , a knowledge of  $\nu_l(z)$  at *finite* frequency in which case the imaginary part of  $\nu_l(z)$  will produce a *shift* in the plasma oscillation frequency. This shift clearly depends on the plasma parameter through Eq. (3). Before analyzing the dispersion equation in more detail let us define the *two* characteristic frequencies which enter Eq. (2). The mean-field plasma frequency  $\Omega_p$  is defined as usual by

$$\Omega_p^2 = \sum_{\sigma=1,2} \omega_{p,\sigma}^2; \quad (5)$$

$$\omega_{p,\sigma}^2 = 4\pi\rho_{e,\sigma}^2/\rho_{m,\sigma} = 4\pi e_{\sigma}^2 n_{\sigma}/m_{\sigma},$$

where  $\rho_{e,\sigma} = e_{\sigma} n_{\sigma}$  and  $\rho_{m,\sigma} = m_{\sigma} n_{\sigma}$  denote, respectively, the mean charge and mass density of the  $\sigma$  species. The second characteristic frequency,  $\Omega$ , entering Eq. (2) can be called the hydrodynamic plasma frequency as it is defined in terms of the total mean charge and mass density,

$$\rho_e = \sum_{\sigma=1,2} \rho_{e,\sigma} \quad \text{and} \quad \rho_m = \sum_{\sigma=1,2} \rho_{m,\sigma},$$

by

$$\Omega^2 = 4\pi\rho_e^2/\rho_m \equiv \Omega_p^2 - \Omega_0^2. \quad (6)$$

From Eq. (6) we see that  $\Omega$  is always smaller than  $\Omega_p$ , the subsidiary frequency  $\Omega_0$  being defined as

$$\Omega_0^2 = 4\pi\rho_0^2/\rho_m; \quad (7)$$

$$\rho_0^2 = \rho_{m,1}\rho_{m,2}(\rho_{e,1}/\rho_{m,1} - \rho_{e,2}/\rho_{m,2})^2.$$

Let us now analyze the dispersion equation (2). For the TCP we have  $\rho_e = 0$  and hence from Eq. (6),  $\Omega = 0$ . The dispersion equation for the TCP

reads then

$$\Delta_{ni}^{\text{TCP}}(\vec{k}=\vec{0}, z) \equiv z^2[z^2 - \Omega_p^2 + i\nu_i(z)z] = 0. \quad (8)$$

As discussed elsewhere,<sup>6</sup> the two modes corresponding to  $z^2=0$  generate, at small but finite  $k$ , two sound waves. The character of the remaining solutions of Eq. (8) changes profoundly with the value of  $\nu_i(z)/z$ . For strong coupling I expect  $|\nu_i(z)/z| \gg 1$  and Eq. (8) leads then to an interspecies momentum relaxation mode and an *ap-proximate* hydrodynamical mode describing a

charge conduction and diffusion process in agreement with an earlier finding.<sup>8,6</sup> For weak coupling,  $|\nu_i(z)/z| \ll 1$ , these solutions describe instead weakly damped plasma oscillations at the complex frequency  $z = \pm \Omega_{\text{TCP}} - \frac{1}{2}i\Gamma_{\text{TCP}}$

$$\Omega_{\text{TCP}} = \Omega_p \left[ 1 + \frac{1}{2} \nu_i(\Omega_p) / \Omega_p \right]; \quad \Gamma_{\text{TCP}} = \nu_R(\Omega_p), \quad (9)$$

where Eq. (4) has been used. Below I will show that for weak coupling  $\Omega_{\text{TCP}}/\Omega_p > 1$ . Let us, however, first consider the case of the BIM. I rewrite its dispersion equation (2) with the aid of Eq. (6) as

$$\Delta_{ni}^{\text{BIM}}(\vec{k}=\vec{0}, z) \equiv z[z + i\nu_i(z)] \left[ z^2 - \Omega_p^2 + \frac{i\nu_i(z)}{z + i\nu_i(z)} \Omega_0^2 \right] = 0. \quad (10)$$

From Eq. (10) we see that no sound modes will develop in the BIM. At nonvanishing  $k$  the  $z=0$  solution of Eq. (10) generates a genuine hydrodynamic diffusion mode whereas  $z + i\nu_i(z) = 0$  leads to nonhydrodynamical relaxation modes. The remaining solutions of Eq. (10) describe plasma oscillations occurring at the frequency  $\Omega_p$  in the weak-coupling limit ( $|\nu_i(z)/z| \ll 1$ ) and at the lower frequency  $\Omega$  in the strong-coupling limit ( $|\nu_i(z)/z| \gg 1$ ). These limiting frequencies are in agreement with the predictions<sup>1</sup> from mean-field theory ( $\Omega_p$ ) and from phenomenological hydrodynamics ( $\Omega$ ). The solutions of Eq. (10) in the vicinity of these limiting frequencies read, for  $z = \pm \Omega_{\text{BIM}} - \frac{1}{2}i\Gamma_{\text{BIM}}$ ,

$$\Omega_{\text{BIM}} = \Omega_p \left( 1 + \frac{1}{2} \frac{\nu_i(\Omega_p)}{\Omega_p} \frac{\Omega_0^2}{\Omega_p^2} \right), \quad \Gamma_{\text{BIM}} = \nu_R(\Omega_p) \frac{\Omega_0^2}{\Omega_p^2}, \quad (11)$$

for weak coupling,  $|\nu_i(\Omega_p)/\Omega_p| < 1$ ; and

$$\Omega_{\text{BIM}} = \Omega \left( 1 - \frac{1}{2} \frac{\nu_i(\Omega)}{\Omega} \frac{\Omega_0^2}{|\nu_i(\Omega)|^2} \right), \quad \Gamma_{\text{BIM}} = \nu_R(\Omega) \frac{\Omega_0^2}{|\nu_i(\Omega)|^2}, \quad (12)$$

for strong coupling,  $|\nu_i(\Omega)/\Omega| > 1$ . Notice that in all cases the sign of the frequency shift is determined by the sign of  $\nu_i$ . For the BIM, there is also the exceptional case  $\Omega_0 = 0$  where the plasma oscillations always occur at  $\Omega_p$  and are undamped (at  $k=0$ ) independently of the coupling. This can be understood by observing that when  $\Omega_0 = 0$  the charge and mass density fluctuations are proportional to each other just as in the case of the OCP.<sup>3</sup>

I now proceed with a calculation of  $\nu_i(\omega)$  for weak coupling which indicates that the frequency shift is positive,  $\nu_i(\Omega_p) > 0$  for  $\Gamma_\sigma < 1$  ( $\sigma = 1, 2$ ). This was also the case in the MD experiments<sup>1,7</sup> although these were performed outside the weak-coupling region. For weak coupling we can use the disconnected approximation<sup>9</sup> for the memory function  $\Sigma$  in Eq. (1) and obtain from Eq. (3) to dominant order in  $\Gamma_\sigma$  ( $\sigma = 1, 2$ )

$$\nu_i(\omega) = \frac{8}{3} \int_0^\infty dk \int_0^\infty dt e^{i\omega t} \sum_{\sigma=1,2} (\beta/\rho_{m,\sigma}) [S_{\rho\rho}(k,t)S_{\rho\sigma\rho\sigma}(k,t) - S_{\rho\rho\sigma}(k,t)S_{\rho\sigma\rho}(k,t)], \quad (13)$$

where

$$S_{\rho\rho} = \sum_{\sigma,\sigma'} S_{\rho\sigma\rho\sigma'},$$

$S_{\rho\rho\sigma} = \sum_{\sigma'} S_{\rho\sigma'\rho\sigma}$  while  $S_{\rho\sigma\rho\sigma'}(k,t)$  are the equilibrium space-time correlation functions of the charge densities of species  $\sigma$  and  $\sigma'$ . As a first estimate I evaluate the right-hand side of Eq. (13) analytically in the Landau approximation. Cutting off the divergent  $k$  integral one obtains

$$\nu_i^L(\omega) = \nu_0 \int_{k_{\text{min}}}^{k_{\text{max}}} (dk/k) [1 + i \operatorname{erf}(\omega/2kv)] \exp(-\omega^2/4v^2k^2), \quad (14)$$

where

$$v^2 = \frac{1}{2} \sum_{\sigma=1,2} v_{\sigma}^2, \quad v_{\sigma}^2 = (m_{\sigma}\beta)^{-1},$$

and

$$\nu_0 = \left(\frac{2}{3\pi}\right)^{1/2} \Gamma_1^{3/2} \omega_p \frac{n_2 e_2^2}{n_1 e_1^2} \left(1 + \frac{n_1 m_1}{n_2 m_2}\right) \left(1 + \frac{m_1}{m_2}\right)^{-1/2}.$$

Notice that  $\nu_i^L(\omega=0) = \nu_0 \ln(k_{\max}/k_{\min})$  reduces to the Spitzer value whereas at  $\omega = \Omega_p$  as needed for Eqs. (9) and (11), one can take  $k_{\min} = 0$  in Eq. (14) and obtain to dominant order in  $a = \omega/2vk_{\max} \ll 1$

$$\nu_i^L(\omega) = \nu_0 \left\{ \ln(e^{-2\gamma} a^{-4}) + i \left[ \frac{1}{2\pi} + O(a) \right] \right\}, \quad (15)$$

where  $\gamma$  is Euler's constant. In the particular case of the  $H^+ - He^{++}$  mixture with  $n_1 = n_2$  as considered in the MD experiment<sup>1</sup> one obtains from Eqs. (11) and (15),  $\Omega_{BIM}/\Omega_p = 1 + 0.08\Gamma^{3/2}$  whereas for the TCP considered in Ref. 7 one obtains from Eqs. (9) and (15)  $\Omega_{TCP}/\Omega_p = 1 + 0.26\Gamma^{3/2}$  with in both cases  $\Gamma = 2^{1/3}\Gamma_1$ . Hence, contrary to the result of a sum-rule analysis,<sup>1</sup>  $\Omega_{BIM}$  has been shown here to be temperature dependent ( $\Gamma \sim \beta$ ). A tentative plot of  $\Omega_{BIM}(\Gamma)$  based on Eqs. (11) and (12), with for instance  $\nu_i(\Omega)$  taken as positive also in Eq. (12), and combining the present theoretical and experimental<sup>1</sup> findings is given in Fig. 1. In view of the fact that a measurement of  $\Omega_{BIM}$  could make possible a deduction of the system's temperature, it would be of interest to produce both theoretical and MD values of  $\Omega_{BIM}$  over a wide range of  $\Gamma$  values leading to a better knowledge of  $\Omega_{BIM}(\Gamma)$ .

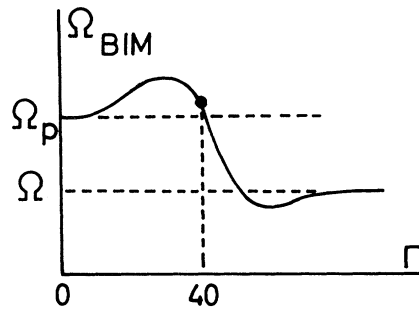


FIG. 1. Tentative plot of  $\Omega_{BIM} = \Omega_{BIM}(\Gamma)$  based on the theoretical findings. The MD value (Ref. 1) is indicated by the dot.

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### Nuclear-Acoustic-Resonance Determination of the Electronic Contribution to the Electric-Field-Gradient Tensor in Molybdenum

E. Fischer,<sup>(a)</sup> V. Müller, D. Ploumbidis, and G. Schanz  
*Fachbereich Physik, Freie Universität Berlin, 1000 Berlin 33, Germany*  
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Nuclear acoustic-resonance absorption and dispersion of both isotopes  $Mo^{95}$  and  $Mo^{97}$  have been observed for the first time in molybdenum. From quantitative evaluation of the nuclear-acoustic-resonance results in the conduction-electron contribution to the tensor  $\vec{S}$  connecting the electric field gradient to elastic strains have been determined at high magnetic fields. Comparison of the electronic contributions to the diagonal and off-diagonal elements of the electric-field gradient in Mo, Ta, and Nb show a disparity in the dependence on the density of states.

Since in cubic metals the electric field gradient (EFG) vanishes at a nuclear site, most of the investigations of EFG have dealt with noncubic metals and alloys. In the presence of an acoustic wave, however, the cubic point symmetry is de-

stroyed and a sound-induced dynamic electric field gradient (DEFG) is created. Hence, nuclear acoustic resonance (NAR) enables one to extend the EFG investigations to cubic metals and alloys. Furthermore, NAR experiments allow some ad-