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Estimating the ion correlation parameter from EXAFS experiments on dense plasmas

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Abstract. The analysis of extended x-ray absorption fine structure (EXAFS) data by Djaoui *et al* led to normalised estimates of the value of Γ, the ion correlation parameter, partly due to the uncertainty in the ionization state of the Al plasma. We adopt a simplified version of the interatomic force model of Laughlin to include the effect of electron screening and show how to estimate the temperature dependence of the Debye–Waller factor for a dense plasma system. This allows the experimental values of the Debye–Waller factor to be used to give limits on both the temperature and, via the Thomas–Fermi ionisation formula of More, on Γ. A subsequent assumption allows estimates of the actual correlation parameter, which are physically reasonable for the experiment in question. We discuss how it will be necessary to develop a model using radial distribution functions to examine properly the question of temperature estimation from EXAFS experiments in the dense plasma regime.

1. Introduction

Much interest has been focused on strongly-coupled plasmas which, at low temperatures of less than a few eV, are typically at least as dense as solid matter. Such systems can be characterised by the ion correlation parameter Γ, the ratio between the potential and kinetic energies of a typical particle defined by

$$\Gamma = (Z_* e)^2 / R_0 k_B T \tag{1}$$

where Z_* is the ionization state, e the electron charge, R_0 the ion-sphere radius and $k_B T$ the temperature in energy units. We shall use atomic units such that $e = m = \hbar = 1$, and temperatures (T) will be measured in Hartrees (1 H = 27.2 eV), distances in Bohrs (1 Bohr = 5.3×10^{-9} cm). A strongly coupled system has $\Gamma > 1$ whereas $\Gamma \ll 1$ is found in classical plasmas, e.g. tokamaks, stellar atmospheres; in dense plasmas we find that the behaviour is dominated by inter-ion Coulombic forces and the motions of ions are strongly correlated (Ichimaru (1982)). Such systems are found in white dwarfs (Van Horn (1980), Schatzman (1980)), giant planetary interiors (Ichimaru (1982)) and in compressed solids. The free electrons in such systems are partially or totally degenerate (Rose (1984), Ichimaru (1982)).

The first direct experimental observation of ion correlation in a dense plasma was reported in experiments using the Vulcan laser at the Rutherford–Appleton Laboratory by the Essex group (Hall *et al* (1988)). These experiments used EXAFS as a diagnostic on a plasma source (Djaoui *et al* (1989), hereafter D89) to infer the density and temperature. The experiments generated a colliding shock on an Al foil of a few microns in width which was then back-lighted by the x-ray continuum from a U source. Compressions of up to a

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few times solid density were obtained and the sample heating amounted to approximately 0.04 H, as detailed in D89.

Extended x-ray absorption fine structure (EXAFS) is the observed modulation of the x-ray absorption coefficient $\mu(E)$ on the high-energy side of an absorption edge, usually the k-edge, and has been studied for many years. Good reviews of this topic are presented by Lee *et al* (1981), Hayes and Boyce (1983), Koningsberger and Prins (1980). The cross section for the photoelectric effect is altered from the solitary atom case by the presence of surrounding atoms, which backscatter the outgoing photoelectron wave and thus produce an interferometric modulation in the probability of absorption of the incoming x-ray. The oscillatory part of μ, $\chi(E)$, is isolated as

$$\chi(E) = (\mu(E) - \mu_0(E))/\mu_0(E) \tag{2}$$

where the background absorption is μ_0, corresponding to a single atomic site. The spectrum obtained contains information about the type and positions of neighbouring shells of atoms (Sayers *et al* (1971), Stern *et al* (1975)), and the sample temperature (Beni and Platzman (1976), Stern *et al* (1975), Rabe *et al* (1979)). The standard theory of EXAFS is well developed (e.g. Gurman (1988), Gurman *et al* (1984)). The simplest EXAFS formula is the plane-wave single-scattering approximation

$$\chi(k) = \sum_j N_j f(k,\pi) \exp(-2R_j/\lambda(k)) \exp(-2\sigma_j^2 k^2)[\sin(2kR_j + \varphi(k)]/kR_j^2 \tag{3}$$

and the wavevector k is related to the photon energy E and threshold energy E_0 by

$$k = 2\sqrt{(E - E_0)} \tag{4}$$

where the sum is taken over the j surrounding atomic shells, at radii R_j and with N_j atoms in each. The backscattering amplitude is $f(k,\pi)$ and the phase shifts are $\varphi(k)$. Inelastic losses are often accounted for by a mean-free path $\lambda(E)$ (Gordon and Djaoui (1992)) and temperature effects can be approximated by the Debye–Waller factor σ_j^2. This gives the mean square relative displacement of atoms in a given shell around the central absorbing atom. The fully correct version of the EXAFS formula which includes spherical waves is given by the Muller and Schaich (1983) prescription, and is valid down to lower energies than the plane wave formula. At low energies multiple scattering may be an important consideration and conventionally the true EXAFS regime does not start until, say, $E - E_0 > 1 - 2$ H above the edge. The high energy data is limited by the increasing temperature and experimental noise, so the window in the k space is limited, often restricting attention to the analysis of the first shell of neighbouring atoms (Lee *et al* (1981), Crozier and Seary (1980)).

2. The Debye–Waller factor

The Debye–Waller factor σ^2 accounts for the thermal atomic motions and EXAFS takes a snapshot of one particular configuration, so an average over such configurations is necessary. If the vibration amplitudes are small, or neighbouring atoms satisfy an effectively Gaussian distribution over distance, then the use of the Debye–Waller factor is justified. (Strictly, we should discuss σ_j^2, where j denotes the jth shell of atoms, but often it is assumed to be the same for all contributing shells; in this particular case, we are concerned with only the first

shell). This is expected to be valid for Al at low T, say, but beyond melting temperature the approaches developed to relate σ^2 to T are expected to be inadequate, where for Al the melting temperature is $933K = 2.95 \times 10^{-3}$ H. In fact, a naive extrapolation of the low-temperature formulae of Beni and Platzman (1976) and Sevillano *et al* (1979) yields unreasonably large values for σ^2 at temperatures around 0.1H and above.

Most theories predict a linear dependence of σ^2 on T for low T, where the coefficient depends on the model used for the density of modes which contribute to the vibrational motion. There is also some arbitrariness in these relationships through their dependence on the Debye or Einstein temperature, and realistic calculations of the density of modes are computationally involved.

Sevillano *et al* (1981) give the following formula for the Debye–Waller factor by using the correlated Debye model of Beni and Platzman (1975)

$$\sigma^2 = 2TM^{-1}\langle\omega^{-2}\rangle \tag{5}$$

for atomic mass M and $\langle\omega^{-2}\rangle$ the inverse second moment of the frequency distribution of modes contributing to the vibrational motion. D89 used this result to estimate Γ via the formula

$$G = 2\omega_p^{-2}\langle\omega^{-2}\rangle R_0^2/3\sigma^2 \tag{6}$$

for the plasma frequency ω_p. As the value of $\langle\omega^{-2}\rangle$ was unknown, it was assumed to be constant over different laser shots and hence a series of 'normalized' values of Γ were found, with only their relative values being of significance. The values obtained are shown in figure 2. This was an inconclusive result from an otherwise successful and innovative experiment. We believe it is possible to gain a better estimate of Γ and provide an upper bound as will be shown below.

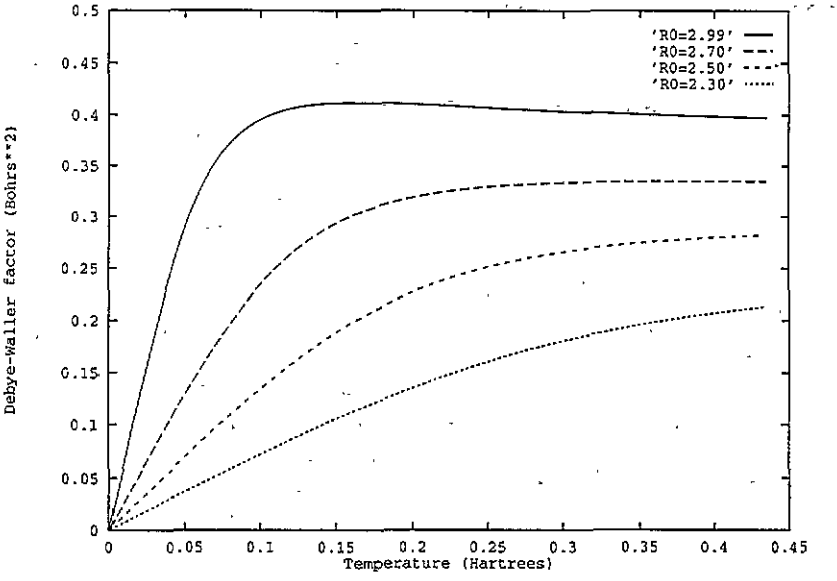

Figure 1. $\sigma^2(T)$ for various densities, specified by the value of cell radius R_0. As the temperature rises, the flattening of the curves shows evidence for the stabilisation effect found by Laughlin (1986).

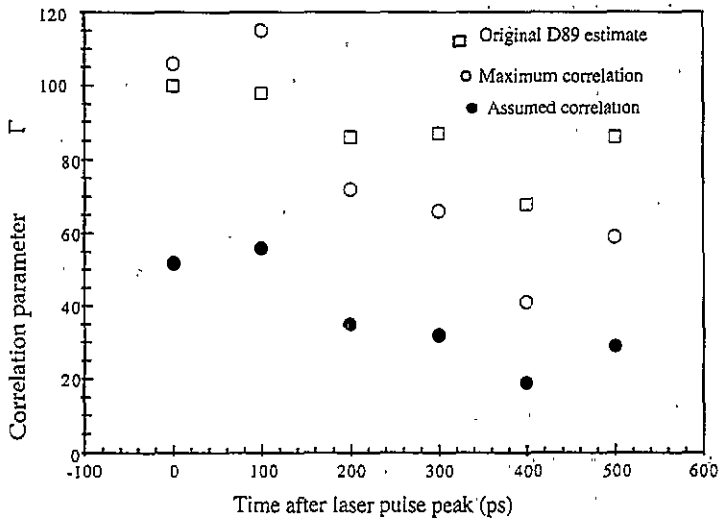

Figure 2. Γ at different times in the experiment relative to the peak of the laser pulse. The values of D89 are given plus the two estimates from this work, from equations (13) and (14).

3. Theory

For temperatures beyond melting, these relations from the correlated Debye model as used by Beni and Platzman (1976), Sevillano *et al* (1979) are not expected to give reliable results. We instead adopt the model of Laughlin (1986) for the temperature dependence of interatomic forces. This is based on Thomas–Fermi theory, which is accurate for high compressions but has known inaccuracies at solid density. A review of Thomas–Fermi theory can be found in Spruch (1991). Some aspects of Laughlin's work are controversial, as some of the predictions of his model seem to be physically questionable, e.g. the non-monotonic behaviour of the Fermi energy as a function of density and temperature. However, in the spirit of Laughlin's paper, we seek qualitative trends and not detailed prescriptions, and expect that his model may give interesting and relevant results for the conditions pertinent to the experiment in question, where we are below the (Fermi) temperatures and where this anomalous behaviour is predicted. We have taken the 1D hydro-code MEDUSA simulations (D89) to give rough estimates of temperature, and the densities are at most a few times those of solids. Laughlin showed how the effects of temperature could be modelled by displacing the ions a distance r_0 away from the centres of spherical cells, radius R_0, and calculated that the interatomic forces were substantially stiffened as the atom reached temperatures between the Fermi level and that for complete ionization, so that the ions start to behave like hard spheres. Laughlin gave expressions for the expectation value of the mean square particle displacement. In fact, the forces are characterised by the r_0 dependence of the electron contribution to the free energy, A. This allows us to treat the ions as if they were attached to springs with a spring constant γ which is given by

$$\gamma = 3Z_*^2/R_0^3 \tag{7}$$

where R_0 is the cell radius and Z_* the ionization state of the atom, i.e. the net charge (positive) of the nucleus plus that of bound electrons. Such a Hooke-law behaviour is valid

for displacements of up to 25% of the cell radius, and this figure increases as the temperature rises. He defined the mean displacement $\bar{r}$ as

$$\bar{r} = C \int_0^{R_0} 4\pi r^3 \exp(-A(r)/T)\,dr \tag{8}$$

with an analogous expression for $\overline{r^2}$, for suitable normalization constant C, and shows that there is a stabilization of the values of $\overline{r^2}$ at temperatures in the range 1–100 Hartrees. We adopt a simplified version of this treatment. First, we take for A, the electron contribution to the total free energy, the expression

$$A = A_0 + \gamma r^2/2 \tag{9}$$

where A_0 is a constant, and use in (7) for the spring constant γ the value of Z_*, which is the value derived from the More formula (1985) for the ionization state. To find the effective spring constant, this must be corrected by a factor that will account for screening of the ionic charges by the free electrons. It has been shown by Djaoui *et al* (1988) that screening by the free electrons acts to reduce the effective value of Z_* and hence the effective correlation parameter by the factors $\exp(-r_s/2)$ and $\exp(-r_s)$, respectively, where r_s is the usual electron Wigner–Seitz radius. The effective values are then Z_*' and Γ' given by

$$Z_*' = Z_* \exp(-r_s/2) \tag{10}$$

$$\Gamma' = \Gamma \exp(-r_s). \tag{11}$$

This was shown by using a variational method based on the Gibbs-Bogoliubov inequality with the one-component plasma (OCP, see below) free energy as a reference system. The screening effect was calculated in the random phase approximation. For different versions of the electronic dielectric function, similar results were obtained. In fact this paper claims to underestimate the screening for densities in the range of interest by omitting a small Γ-dependence, but this omission is at least partially compensated for by our inclusion of varying ionization levels, which was absent from that work. Thus expressions for $\bar{r}$ and $\overline{r^2}$ can be found which combine to give the mean square displacement for a single atom, σ_0^2, as

$$\overline{r^2} - \bar{r}^2 = \sigma_0^2 \tag{12}$$

adopting the notation of Stern *et al* (1975) for this quantity. This is plotted as a function of temperature in atomic units (Hartrees and Bohrs) in figure 1 for a range of densities, parameterised by the differing cell radii R_0. To get the real EXAFS Debye–Waller factor requires the relative mean squared displacement for a given shell of atoms, σ_1^2, and this is related to σ_0, depending on the degree of correlation. As pointed out by Stern *et al* (1975), a completely uncorrelated system will have

$$\sigma_1^2 = 2\sigma_0^2 \tag{13}$$

but a perfectly correlated case will give all vibrations in phase so that $\sigma_1^2 = 0$. As the degree of such correlation is unknown a priori, we adopt the simplifying assumption that

$$\sigma_1^2 = \sigma_0^2 \tag{14}$$

with its known temperature dependence. This assumption will be examined retrospectively below, but allows for partially correlated atomic motions, in a way similar to the results of Stern *et al* (1975) for metallic Cu. By using equation (13) we find upper limits for the correlation parameter Γ, and from (14) a proposed improvement on the previous estimates. We see that the relation is approximately linear over the energy range of 0–0.06 Hartrees, which should be adequate for the EXAFS experiments, where the Medusa simulations showed peak temperatures of about 0.04 H, and this is liable to be an overestimate as Medusa is a 1D code designed primarily for other regimes of work.

4. Application to results

We have found that an observed, or strictly speaking, fitted-to-observations, σ^2 can be identified with a temperature T. The fitting procedures are described elsewhere (D89) and can be tackled using the Daresbury EXCURV package (Binsted *et al* (1990). Taking the data from D89 we match the values of σ^2 to temperature from a calculated table. The More formula gives an ionization state, and hence the effective Z_*' and Γ' can be calculated. The results are shown in table 1 and figure 2, giving the values of the correlation parameter from using both (13) and (14). We see that the maximum values of Γ are not unreasonable compared to those tabulated in Djaoui *et al* (1988), and the degree of correlation peaks as expected some time after the laser pulses collide, and thereafter it tends to decrease as the plasma cools and expands freely. The uncertainties in these values are similar to those in D89, namely a relative error of 20-25% in the size of Γ.

Table 1. Estimated values of correlation coefficient using both equation (12) and (13). The experimental values of σ^2 come from D89, Z_* is found from More's formula for the Thomas–Fermi ionisation state.

$t(ps)$	R_1	$\sigma^2(B^2)$	$T(H)$	Z^*	Γ'	Γ
$\sigma^2 = 2\sigma_0^2$ values						
0.0	2.83	0.18	0.023	0.93	13.5	106
100	2.93	0.19	0.018	0.85	13.3	115
200	2.62	0.18	0.044	1.16	11.6	72
300	2.50	0.16	0.057	1.30	11.8	66
400	2.26	0.17	0.131	1.66	9.3	41
500	2.42	0.15	0.072	1.41	11.6	59
$\sigma^2 = 2\sigma_0^2$ values						
100	2.83	0.18	0.047	0.95	6.7	52
200	2.93	0.19	0.038	0.86	6.6	56
300	2.62	0.18	0.093	1.18	5.7	35
400	2.50	0.16	0.12	1.33	5.8	32
500	2.26	0.17	0.30	1.75	4.4	19
600	2.42	0.15	0.15	1.45	5.7	29

5. Discussion

The values of Γ inferred are physically reasonable: a benchmark can be taken by considering the OCP model, whose properties are dependent solely on the value of Γ. In the one-component plasma (OCP) the ions are point charges and the electrons are replaced by a

uniform charged fluid with no effective properties normally associated with a real electron gas (screening, correlation). The OCP is expected to be a good model at ultra-high density in, for example, a white dwarf interior. Extensive simulations of the OCP have made it one of the best-studied physical systems, in numerical terms. In a series of papers by Hansen (1973), Slattery *et al* (1980) and DeWitt and Rosenfeld (1979), it has been shown how the OCP freezes into a bcc lattice at a value of $\Gamma = 178 \pm 1$, and there is a further transition to the fcc lattice at $\Gamma = 192 \pm 1$. Although a real Al plasma will be subject to screening and to the quantum behaviour of electrons, it can be expected that these values are upper limits for the expected values of Γ' that might be observed. In fact Djaoui *et al* show how for solid Al at its melting temperature of 933 K $= 2.95 \times 10^{-3}$ H and $\Gamma' = 156$, not too dissimilar from the OCP value.

A source of error in this approach is the uncertainty in relating σ_1^2 to σ_0^2, as the choice of a proportionality constant will strictly depend on the degree of correlation (which is unknown a priori), plus the use of the Thomas–Fermi ionization formula which does not include any of the possible influences from the low-energy electron 'scattering resonances' discussed by More (1985), in particular their effect on screening. We feel justified in adopting the simple relation of equation (14) in the absence of a more detailed prescription for this choice.

For realistic modelling of EXAFS spectra in the density–temperature regime accessible to laser-compressed experiments, it will become necessary to replace the 'sum over shells' approach with an integral over the radial distribution function (RDF) for the plasma, which will properly account for the actual positions of neighbouring ions. A preliminary work along these lines will be presented in a future publication, showing that the usefulness of the Debye–Waller factor breaks down as the shell structure becomes less distinct. The problem shifts to the accurate determination of $g(r)$, the RDF appropriate at a given density and temperature. Even before this higher temperature range is reached, there is a danger of inferring incorrect parameters from EXAFS analysis which asumes that the forces contributing to vibrational motion are essentially harmonic—Eisenberger and Brown (1979), Crozier and Seary (1980) have discussed this problem and shown that incorrect and misleading conclusions can be made even at modest temperatures when anharmonicities are present—these effects are more important as the temperature rises and the hard-core repulsion is felt.

Another potential approach is that using cumulant analyses (Bunker (1983) and Crozier *et al* (1980)) which allow determination of structural parameters when the assumption of harmonic potentials is not guaranteed, and provides tests for the detection of asymmetric distribution functions.

6. Conclusion

We have used the data from EXAFS experiments on dense Al plasma to estimate the temperature and correlation factor using a new method which is more appropriate to the density and temperature regime expected. The answers obtained have more physical significance than the previous estimates of D89 since a model from Laughlin has been used for the interatomic forces and the effects of screening by the free electrons have been approximately accounted for. Although there are aspects of Laughlin's work that are controversial, we believe that our regime of interest does not overlap with that where these uncertainties apply, as the temperatures are below the Fermi temperature throughout. In addition we have used a reliable ionization model which models the temperature and

density dependence of Z_*. An upper limit for Γ is suggested. Further progress to obtaining diagnostic information from such experiments at higher temperature will require the use of a plasma radial distribution function when in regimes where the use of the Debye–Waller factor is no longer physically justified.

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