Instability of Electron Gas in an Expanding Metal

The properties of dilute electron gas have attracted attention since Wigner predicted that it crystallizes in its dilute limit [1]. The behavior of electron gas is often discussed in terms of its ground-state energy, given as a function of the expansion parameter r_s (the Wigner-Seitz radius in units of the Bohr radius). r_s increases with decreasing density and scales approximately as the ratio of the electron interaction energy to the average kinetic energy. As the interaction energy dominates the kinetic energy (r_s increasing), various phase transitions or instabilities such as ferromagnetism or superconductivity have been predicted. Among the various properties of electron gas, compressibility is an intriguing property deduced from the ground-state energy. It becomes negative when $r_s > 5.25$ [2], implying an instability in low-density electron gas. Theoretically, a negative compressibility results in a negative static dielectric function (DF) of the electron gas, $\varepsilon(\mathbf{q}, 0)$, for small $|\mathbf{q}|$. This has interesting consequences, including the possibility that test charges of the same sign might attract.   However, the existence of a negative DF in a three-dimensional system has not yet been confirmed.

Expanded fluid alkali metals [3] are good systems for investigating the problem of electron gas instability for several reasons. First, alkali metals are a natural realization of the electron gas because of the nearly spherical Fermi surface of the conducting electrons. Second, a continuous and substantial reduction in electron density is possible by volume expansion along the liquid-vapor saturation line as shown in Fig. 1. Third, positive ions in fluids can readjust their positions more easily than those in solids; thus structural changes might be clearer in fluids through the coupling of electrons and ions.

We have recently fabricated a sample cell with X-ray windows made of single-crystalline molybdenum, which is resistant to the high reactivity of hot alkali metals. By combining this cell with a high-pressure apparatus, we have succeeded in measuring the X-ray diffraction and small-angle X-ray scattering (SAXS) of fluid rubidium over a wide range of temperatures and pressures from the triple point up to supercritical regions [4]. X-ray diffraction measurements were carried out in a dispersive mode at beamline **BL28B2** using an incident white beam and an energy resolving germanium detector. Small-angle X-ray scattering measurements were performed at beamline **BL04B2** using monochromatic 38 keV X-rays, with the scattered X-rays detected using an imaging plate. For the first time, stable and precise measurements were carried out from the triple point up to 2123 K and 22.0 MPa, beyond the critical point of rubidium.

The variation of the position of the first maximum, R_1, is shown as a function of density in Fig. 2(a). R_1 corresponds to the nearest-neighbor distance and it is located at approximately 4.85 Å at 373 K. It decreases slowly with decreasing density to about 1.1 g·cm^{-3}, and then drops more rapidly. In Fig. 2(b), the density dependence of the S(0) of fluid Rb is shown. Below the density of 1.1 g·cm^{-3}, S(0) starts to increase substantially with decreasing density, which indicates the appearance of spatial atomic fluctuations in the fluid.
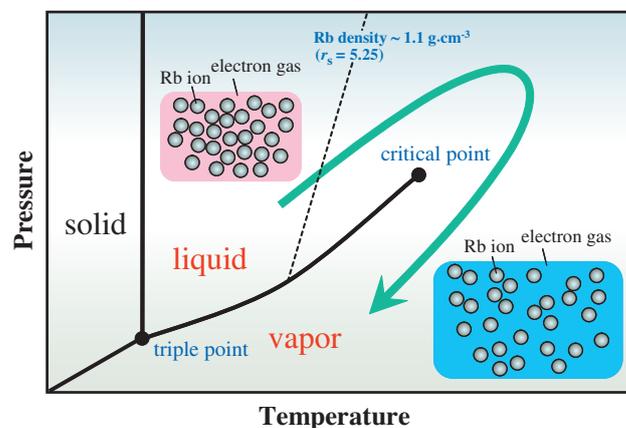
The change in character of the SAXS results

Fig. 1. Schematic illustration of the phase diagram of rubidium (T_c = 2017 K, P_c = 12.45 MPa) [3].   A continuous and substantial reduction in *electron* and ion density is possible by volume expansion along the liquid-vapor saturation line.
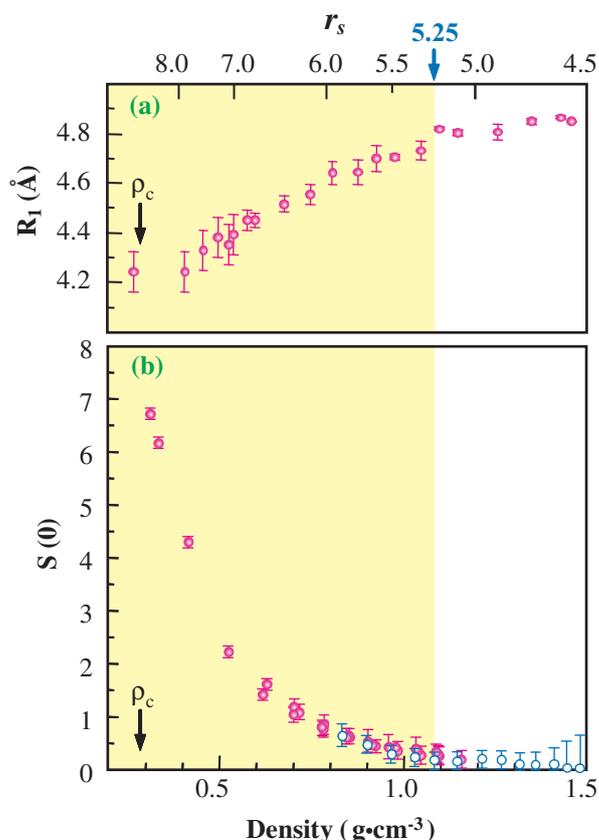
Fig. 2. Density dependence of the structural parameters of fluid rubidium. **(a)** Density dependence of interatomic distance R_1 and **(b)** density dependence of S(0). Dots represent the data measured from 373 K - 1.0 MPa up to 2123 K - 22.0 MPa along the liquid-vapor saturation line. Open circles indicate the data measured from 373 K up to the point near the saturation line at a constant pressure of 5 MPa. The critical density is denoted by ρ_c. The corresponding scale of r_s is shown on the upper axis of the graph of R_1.

occurs at a density, r_s, of between 5 and 5.5, near the critical value of r_s (= 5.25), beyond which the static electron DF becomes negative [2]. The negative electron DF generates an attractive Coulomb interaction among test charges with the same sign. Therefore, the structural features of expanded fluid Rb, such as local contraction and the appearance of the density fluctuation in the metallic state below 1.1 g·cm^{-3}, might be caused by the enhancement in the attractive interaction among the ions in the medium of the electron gas with a negative DF. This also suggests a possible connection to electron pairing. A close connection between the negative static DF and superconductivity has been suggested [5] in the doped cuprates [6]. Expanded fluid alkali metals may show superconductivity at low temperatures if their structural response to the negative electron DF can be frozen.

Kazuhiro Matsuda [a,*], Kozaburo Tamura [a] and Masanori Inui [b]

[a] Graduate School of Engineering, Kyoto University
[b] Graduate School of Integrated Arts and Sciences, Hiroshima University

*E-mail: kazuhiro-matsuda@mtl.kyoto-u.ac.jp

References
[1] E.P. Wigner: Phys. Rev. **46** (1934) 1002.
[2] G. Giuliani and G. Vignale: Quantum Theory of the Electron Liquid (Cambridge University Press, Cambridge, England, 2005).
[3] F. Hensel and W.W.Warren Jr.: Fluid Metals, The Liquid-Vapor Transition of Metals (Princeton University, Princeton, NJ, 1999).
[4] *K. Matsuda, K. Tamura and M. Inui: Phys. Rev. Lett. **98** (2007) 096401.*
[5] O.V. Dolgov *et al.*: Rev. Mod. Phys. **53** (1981) 81; O.V. Dolgov and E. G. Maksimov: Sov. Phys. Usp. **25** (1982) 688.
[6] M. Tachiki *et al.*: Phys. Rev. B **67** (2003) 174506.