Compton Scattering Measurement to Detect Momentum Distribution of Electrons in Warm Dense Matter

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Detailed study of electrons in warm dense matter is a key to understanding material properties. Up to now there are many methods to detect electron spectral features with optical lasers and x-rays. Here we propose and demonstrate a new measurement method using inelastic Compton scattering of hard x-ray probe radiation. By using a high energy photon beam, above 100 keV, the change of the electron momentum distribution is observed through a change of the scattered x-ray spectrum even in a solid density plasmas.

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At warm dense matter conditions, material macroscopic behavior is sometimes dramatically different from the solid and ideal plasma even though the densitytemperature parameters of warm dense matter are located in an intermediate range between condensed matter and plasma. Up to now, strong reduction of electron conductivity is clearly observed in several experiments [1,2] and predicted in simulations [3]. To understand this phenomenon, localization of electrons is a key idea. In this paper, a simple scheme is considered for study of the transition of material from the solid to a lower density WDM plasma by heating with a laser. As is well known, some electrons in a normal metal are located in the conduction band and have an important role for electronic conduction and are responsible for good reflectivity of visible and infrared light. The other electrons in the materials are bound inside atoms and cannot move freely. In an ideal plasma, outer electrons are ionized from atoms and these free electrons are responsible for electronic conduction. Due to total charge neutrality, in the plasma, the free electron charge density is balanced by an equal charge density of positive ions. Of course there is Coulomb interaction between the ions and electrons and sometimes they recombine and ionize in the normal equilibrium manner. But in the warm dense matter region, electrons sometimes behave as free electron and sometimes they are localized on atoms so that they don't contribute to the conduction.

This localization process is common physics in many materials. But in warm dense matter, it is difficult to study because of the relatively high temperature and short life-time caused by the high pressures. Typical density and temperature parameters of the warm dense matter discussed in this paper are 1/10 solid density and 1 eV temperature. A WDM plasma with these parameters is easy to produce with lasers and occurs in many laser applications.

To measure the electron spectral features in warm dense matter, there are high expectations for Thomson scattering because it can determine temperature and density simultaneously [4]. The spectrum of this scattering is affected by coupling between the electron and the ion. Intense x-ray sources such as a kJ laser produced plasma or x-ray free electron laser pulse are used to get spectrum in a single shot experiment. The photon energy used is about 10~20 keV but a high flux is needed so that the possible sources are limited. The actual spectrum consists of several components and theoretical models are needed to analyze it. Due to the short penetration depth of 10's keV x-rays, measurable thickness of sample is as thin as 10 µm.

Here, we propose a new method to observe the electrons in warm dense matter. A schematic drawing of this measurement is shown in Fig. 1. The measurement is based



Fig. 1 Measurement scheme of inelastic Compton scattering of plasma.

on Compton scattering of high energy photons. The parameters of electrons and photons are related by the momentum and energy conservation law as in the following well-known equations [5].

$$\hbar\omega_1 + \frac{P_1^2}{2m} + U_1 = \hbar\omega_2 + \frac{P_2^2}{2m} + U_2, \tag{1}$$

$$\hbar k_1 + P_1 = \hbar k_2 + P_2, \tag{2}$$

$$\hbar\omega_2 - \hbar\omega_1 + \frac{\hbar^2 |K|^2}{2m} = -\frac{\hbar K \cdot P_1}{2m},\tag{3}$$

here, *m* is electron mass, $\hbar\omega_i$, P_i , U_i , k_i are photon energy, electron momenta, electron potential energies, and photon momenta. The suffix *i* labels parameters before the photon scattering (*i* = 1) and after (*i* = 2). The vector *K* is defined as the difference between incident and scattered photon momenta (k_1 – k_2). In our method, we use hard x-ray beams having photon energy larger than 100 keV. With such high photon energy, the interaction between electrons and x-rays is simple. Normally we assume the impulse approximation in which potential energy of electron is not changed in the collision event so that $U_1 = U_2$. The parameters on the right hand in Eq. (3) are decided and measured in the experimental condition and measurements. Therefore, we determine the initial momentum of the scattered electron as a projection on the K vector.

The merit of the high energy x-ray beam is that we can use the impulse approximation so that any electrons inside plasma, transient material, and solid have the same response without any effect of bonding to the atoms. In addition, the penetration depth for x-rays above 100 keV is more than several mm. We can study a much deeper region in dense target material. This situation is closer to the real configuration of high power laser applications such as laser welding and laser cutting of steels and other metallic materials.

When we consider the generation of warm dense matter from a solid with lasers, the transition process is the following: (1) Cold condensed matter is heated by the laser heat flux. (2) Above the melting and boiling temperatures, the material starts to expand and its temperature increases with the heat flux. (3) The material is partially ionized (WDM) while keeping high density. (4) Expanded material is still heated by incident laser to make a plasma. (5) After large expansion, the density drops below the critical density for the laser. With enough expansion cooling or cooling due to ambient gas, the expanded target material starts to re-condense to produce gas, clusters or droplets. We are interested in the transient process between (2) and (3). As is well known, the electrons inside the solid metal are divided into two components: free electrons in the conduction band and valence electrons bound in atoms. The free electrons are located in the Fermi distribution and have a maximum velocity decided by

$$v_F = \left(\frac{\hbar}{m}\right) \left(\frac{3\pi^2 N}{V}\right)^{1/3},\tag{4}$$



Fig. 2 Calculated spectrum of Compton scattering for two different electron velocity distribution function. The lower two spectrum denotes the spectrum of Fermi and M-B distribution. The upper curve is subtract results the lower two spectrums.

Here \hbar is the Plank constant divided by 2π , N/V is density of conduction electrons. If we assume a spherical Fermi surface, P_1 in Eq. (3) has a symmetrical distribution in momentum space with this velocity in the case of non-crystal metals such as stainless steel.

In the WDM region, we expect the free electrons coming from ionization to be non-degenerate. In this case, as a first approximation, we can use the Maxwell-Boltzmann distribution (M-B), in which velocity distribution f(v) is given by

$$f(v) \, \mathrm{d}v_x \mathrm{d}v_y \mathrm{d}v_z = \left(\frac{m}{2\pi kT}\right)^{3/2} \exp\left(-\frac{m(v_x^2 + v_y^2 + v_z^2)}{2kT}\right) \mathrm{d}v_x \mathrm{d}v_y \mathrm{d}v_z,$$
(5)

here dv_x , dv_y , dv_z are velocity differentials for each direction and kT is temperature. In the WDM region, we expect that some of the electrons in the original Fermi distribution change to this M-B distribution. The percentage of such ionization is estimated from simple Saha equilibrium. This number changes dramatically from 10^{-5} to 1 when the temperature rises from 0.3 to 1 eV for steel.

The expected spectra of Compton scattering for both electrons in the Fermi distribution in the metal and free electrons are calculated by integration of Eq. (3) over the velocity distributions. Typical spectra of these conditions are shown in Fig. 2; in this case the selected temperature is 0.5 eV. The two spectra are clearly different. The horizontal energy axis is the shift of the peak photon energy of the Compton scattering. Normally, the peak position of the Compton spectrum is lower by tens eV from the incident x-ray energy. In Fig. 2 difference between these two spectra is also shown. We see the difference curve has a wide negative component and narrow positive peak. As we men-



Fig. 3 Schematic drawing of inelastic Compton scattering measurement with CW high power laser. The inserted photograph is shadow graph picture for height level controller. The probe areas of >100 keV x-ray are also shown in this picture.

tioned above, all electrons in the material can scatter the incident x-rays in the same manner so that the other tens of electrons per atom located in bound core levels make a spectrum at the same peak position. Therefore, the differential signal rides on a large relatively broad spectrum. To get clean signal for the difference, we need to increase total number of scattered x rays, carefully.

To get proof of principle of this measurement technique, we measured the spectrum of Compton scattering of laser heated steel with a 116 keV photon probe beam. The experimental setup is shown in Fig. 3. The sources of such high energy photon are limited and we use the SPring-8 synchrotron BL08W beam [6]. The beam is shaped by a slit and the beam cross section is $50 \times 200 \,\mu\text{m}$. This beam arrives in pulses with tens of MHz repetition rate and the nominal photon flux is 5×10^9 /s. For production of the WDM condition, we use a high average power fiber laser which has 400 W maximum CW power in a single transverse mode. The laser is focused on the surface of the sample with an f = 200 mm lens and the focal spot diameter is about 50 µm in this experiment. With this irradiation, the sample is melted and evaporated and plasma production occurs. To keep a fixed interaction point of the laser and sample material, we use a cylinder target with diameter of 100 mm and length of 500 mm. When we select proper rotation speed and translation speed along z axis, the interaction point is fixed at the same position. To prevent heating of the measurement system and thermal expansion of optics and sample rotator system, water cooling is used. This target rotation and translation system and the head of the fiber stands on one optical table. The height level is always monitored and fixed with a laser shadowgraph (inserted photograph in Fig. 3) and a vertical translation stage. Probing x-rays enter the interaction area and by changing the height of the optical table, the x-ray probing



Fig. 4 Measured Compton spectrum (below) and the subtract result between that of cold material and that of laser heated expansion material. In the subtract data, the similar spectral response to the theoretical prediction is shown clearly.

area is changed with a $20 \sim 50 \,\mu\text{m}$ step. The x ray probe (> $100 \,\text{keV}$) measures the electrons inside material from solid to the low density plasmas. The observation point of x ray scattering is also limited with another W slit located at the line of sight of the scattered x-rays. This slit width also decides resolution of momentum of electrons because variation of scattering angle in projection term of $K \cdot P_1$ in Eq. (3). As a detector of the > $100 \,\text{keV}$ scattered photons with enough high spectral resolution, we use Liq. Nitrogen cooled semiconductor detector with height level discriminator. We keep the measured photon number of the scattering x-rays less than one for one event of scattering. Typical energy resolution of this system is about 0.5 keV at 116 keV photon energy.

To get difference between electrons in condensed metal matter and WDM plasma, we measure the spectrum as a function of the height above the sample original surface. We get the scattering spectra as a function of position of the x-ray probe inside the plasma. The count of the Compton scattering signal is constant inside solid and decreases in the warm dense matter and plasma region. This information lets us know the effective density in the interaction area. Typically it takes 10 min. to get a clean scattering spectrum at one position. Therefore, to get spectra at about five different positions, all the target cylinder surface is used and should be replaced.

A typical differential spectrum of a stainless steel 316 target is shown in Fig. 4. The lower curve denotes the total Compton scattering spectrum $F_{\text{cold}}(h\nu)$ from cold metallic material, while the upper curve is the difference between scattered signals of WDM region $F_{\text{WDM}}(h\nu)$ and normalized spectrum $\overline{F_{\text{cold}}(h\nu)}$ of the cold material. (This normalization is $\overline{F_{\text{cold}}(h\nu)} = F_{\text{cold}(h\nu)} \cdot \frac{\int F_{\text{WDM}}(h\nu)dh\nu}{\int \int F_{\text{cold}}(h\nu)dh\nu}$.) The peak dif-

ference between $F_{\text{cold}}(h\nu)$ and $F_{\text{WDM}}(h\nu)$ is about 0.5% so that their shapes of the total scattered signal look similar in the scale of the lower spectrum of Fig. 4. We clearly see the differential spectrum is similar to that predicted in the theory (Fig. 2). With comparison to the estimated curve from theory, the temperature of this measured area is about 0.5 eV. The sum number of the ion and neutral atom density in probing area is estimated from the total Compton scattering intensity because that is proportional to total number of the electrons both of bound and free states. The density is estimated to be $1/4 \sim 1/2$ of cold solid density with assumption of 1D expansion. According to the Saha equilibrium, ionization at this condition is about 1%. These parameters are consistent with the warm dense matter region. It is the first experimental result of inelastic Compton measurement for WDM studies.

We propose and demonstrate a new x ray scattering measurement for warm dense matter. We use a pulsed x-ray beam above 100 keV to measure electron momenta in the high-density region of interaction between a high power CW laser and metallic material. After getting the Compton spectrum, subtraction between scattered spectrum of WDM region and that of the cold material is calculated. This differential spectrum agrees with the predicted spectrum, which is a difference of the Compton scattering spectra of free thermal electrons and that for electrons on the Fermi surface of the metallic material. This is a first proof of principle experiment for this new x-ray spectroscopic measurement for WDM condition. Up to now, we use simple assumption of symmetrical Fermi surface in the metal and M-B distribution in plasma. However, we expect discrepancies in the experimental data from this simple assumption. That components will be also important to study WDM detail physics. We need better S/N measured experimental results after longer data accumulation. It will be reported soon.

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