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Theoretical studies on the photoionization cross-sections of solid silver*

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An alternative expression for photoionization cross-section of atoms or molecules and a dielectric influence function (DIF) in a high-density system proposed recently are used to study the photoionization cross-sections of solid silver. It is suggested that a density turning point (DTP) of a photoionized system may be viewed as the critical point where the photoionization properties of atoms in a real system may have a notable change. The results show that the present theoretical photoionization cross-sections are in good agreement with the experimental results of a silver crystal both in structure and in magnitude.

Keywords: photoionization cross-section, solid silver, dielectric constant
PACC: 3280F, 3280, 3380E

1. Introduction

Photoionization and photodetachment processes of isolated atoms or molecules (in ideal gas) are the simplest electron transfer reactions induced by photoabsorption and have been extensively studied for several decades.[1−8] Understanding these microscopic processes and the related properties is fundamental and important to many physical and chemical studies. Photoionization processes of condensed systems (non-ideal case) have received much attention recently[9−13] because of their increasing importance. For example, photoionization data are the necessary inputs for modelling phenomena in plasmas and neutral fluids,[9] etc. Many photoionization experiments have aimed at studying the characteristics and properties of solid-states, oligomers and clusters,[10] the temperature dependence of photoionization cross-sections,[11] the dynamic processes of photoionization of liquids,[12] and the atomic/molecular interactions of photoionization processes,[13] rather than studying them just for a single free atom (or ideal gas). Phenomena such as line broadening and spectral shifts of the optical spectra of atoms in liquid systems contain information on both the dynamics of liquids and the interatomic interactions, which can be studied using interphase theory.[14] Many measured microscopic properties of atomic and molecular photoionization stem from experiments in condensed phases. Photoionization cross-sections of atoms/molecules in condensed phases may be sensitive to the "environmental" effects — the electromagnetic effects of surrounding particles and the interactions among particles. Understanding the relation between macroscopic electromagnetic responses observed in experiments and microscopic properties such as photoionization cross-sections and identifying the influence of the state of aggregation (the number density effects, etc) on the observable microscopic properties still present a challenge.[15]

Generally there exist two main complementary approaches to solving this problem: (i) interphase theory and (ii) cluster physics and chemistry.[14] In the former approach, a given reaction or property is studied as a function of density and properties of the medium in which it occurs from a low-density gas to a liquid or solid. In the present work we investigate the number density effect on photoionization cross-sections of atoms in a real system using the general expression for photoionization cross-sections and the

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dielectric influence function proposed recently.[16] The basic idea of these expressions is that the photoionization cross-sections of atoms in a real system can be described as the coupling between quantum quantity (QQ) and classical quantity (CQ) parts. The QQ part represents the photoionization cross-sections of an isolated particle, while the CQ part may represent most of the important influences of the macroscopic effects on the photoionization cross-sections. The theoretical expression for photoionization cross-sections of condensed matters is based on the experimental Beer–Lambert's law.[17] And the expression is derived directly from the physical response of a photoionized system and is independent of any quantum mechanical method. The theoretical photoionization cross-sections of solid silver are studied using the new expression and compared with the experimental values of both an ideal gas and a solid phase. The interatomic interactions are considered using the well-known Clausius–Mossotti model[18] in this study. It is suggested that a density turning point (DTP) of a photoionized system may be viewed as the critical point where the photoionization properties of atoms in a silver system may have a notable change. The results show that the suggested method is reliable and would open a new way to calculate the photoionization cross-sections of atoms in condensed systems.

The general expression for photoionization cross-sections of a condensed system and the dielectric influence function (DIF) are presented in Section 2. Section 3 applies the new formula and the DIF to solid silver, and analyses the variations of the complex dielectric constant ϵ, the DIF and the photoionization cross sections σ with photon energy ω and number density N respectively. Finally, Section 4 presents our conclusions and discussion.

2. Photoionization cross-sections of condensed phase

The photoionization cross-sections of atoms are usually determined by detecting the light attenuation through a target. It is known from the Beer–Lambert's law that the number density of the medium significantly affects the measured cross-sections. Based on the Beer–Lambert's law, and using the first order approximation in a series expansion on the dielectric constant $\epsilon^{1/2}$ in an ideal gas model, Fano and Cooper[17] gave the expression for photoionization cross-sections σ_i of an isolated atom or molecule used in most theoretical studies,[1−8]

$$\sigma_{\rm i}(\omega) = \frac{4\pi\omega}{c}\zeta, \tag{1}$$

where c is the speed of light, and ζ is the imaginary part of dynamic dipole polarizability $\alpha(\eta + i\zeta)$ of a free atom/molecule. σ_i in Eq.(1) is independent of any macroscopic quantity of a system, and only represents the property of an isolated atom. Equation (1) is valid usually in a range of room temperature (273K) and pressure (1atm) under which a typical number density of particles $N = 3.99\times10^{-6}a_0^{-3} = 2.70\times10^{7}\mu\mathrm{m}^{-3}$ (a_0, the Bohr radius) for an ideal gas and the interatomic interactions among the atoms in an ideal-gas system can be completely neglected.

However, as mentioned in Section 1, photoionization cross-sections of atoms in a condensed system may be sensitive to "environmental" effect which might be represented by some macroscopic properties such as, the complex dielectric constant ϵ of the system. Using the Born–Oppenheimer approximation, Lax suggested an experiential formula for the photoionization cross-sections which is corrected by the dielectric property and electric field of a photoionized system,[19]

$$\sigma(\omega) = \left[\frac{n}{\epsilon}\left(\frac{E_{\rm e}}{E}\right)^2\right]\frac{8\pi^3\omega}{3c}I_{\rm ba}(\omega), \tag{2}$$

where $I_{\rm ba}(\omega)$ represents the square of the matrix element of emission transition from the initial quantum state b to the final state a, ω is the photon energy, n is the index of refraction, ϵ is the dielectric constant of the condensed system, and the factor $(E_{\rm e}/E)^2$ describes the fact that the emitted electron is located in the medium of which the effective field $E_{\rm e}$ may differ from the average macroscopic electric field E. The factor $(8\pi^3\omega/3c)I_{\rm ba}(\omega)$ is an alternative expression for photoionization cross-section σ_i of an isolated atom in Eq.(1), and the factors in the brackets take account of the modification of the electromagnetic field in the crystal to the cross sections. Can the photoionization cross-sections of atoms and molecules in a condensed system be theoretically related to some macroscopic quantity of the photoionization system? Is there a proper and correct expression that describes the macroscopic medium influence of a real system on the photoionization cross-sections? Recently, an alternative expression for photoionization cross-sections of atoms in a real system has been proposed based on the experimental Beer–Lambert's law without using any

mathematical approximation and physical model,[16]

$$\sigma(\omega) = \frac{\sqrt{2}\omega}{Nc}\sqrt{\sqrt{\epsilon'^2 + \epsilon''^2} - \epsilon'}, \tag{3}$$

which is a function of the number density N of particles of the condensed system and can be rewritten in a series expansion as

$$\sigma(\omega) = \frac{\omega\epsilon''}{Nc} f(\epsilon', \epsilon''), \tag{4}$$

$$\begin{aligned} f(\epsilon', \epsilon'') =& \frac{\sqrt{2}}{\epsilon''}\sqrt{\sqrt{\epsilon'^2 + \epsilon''^2} - \epsilon'} \\ =& \frac{1}{\sqrt{\epsilon'}}\left[1 + \sum_{m=2}^{\infty}(-1)^{m+1}\frac{2(2m-3)!!}{(2m)!!}\left(\frac{\epsilon''^2}{\epsilon'^2}\right)^{m-1}\right]^{1/2}, \quad \text{for } \epsilon'' < \epsilon' \end{aligned} \tag{5}$$

ϵ' and ϵ'' are the real part and the imaginary part of the complex dielectric constant $\epsilon(= \epsilon' + i\epsilon'')$ of a macrosystem respectively. The series expansion in Eq.(5) can be used only for the cases $\epsilon' > 0$ and $\epsilon'' < \epsilon'$. Comparing Eqs.(1) and (3), one can see that Eq.(3) is a function of macroscopic properties (ϵ, N) of the system and can be used to evaluate the cross-sections of atoms both in condensed phases and ideal gas systems, while Eq.(1) is only a function of the microscopic property, i.e. the imaginary part ζ of the dipole polarizability α of a single particle and can only describe the cross-sections of an ideal-gas system. Equation (3) is a solid expression and gives a convenient way to understand the influence of the macroscopic electromagnetic responses such as the observable dielectric constant (ϵ', ϵ'') on the microscopic quantity $\sigma(\omega)$. On the right-hand side of Eq.(3), the complex dielectric constant (ϵ', ϵ'') represent the statistic average of the macroscopic dielectric property and implicitly includes the dynamic influence of other particles in the environment on the ionized particle. Usually, linking the gaseous and condensed phases of matter requires multidisciplinary and systematic investigations concerning how the microscopic and macroscopic properties of materials are related with each other and how the photoionization processes are changed. A very useful review of these studies is given by Christophorou *et al.*[14]

Equation (3) can also be reexpressed as[16]

$$\sigma(\omega) = f(N, \epsilon', \epsilon''; \zeta)\sigma_i(\omega), \tag{6}$$

where $\sigma_i(\omega)$ is the photoionization cross-sections of an ideal gas, which is expressed in Eq.(1), and the transformation function

$$f(N, \epsilon', \epsilon''; \zeta) = \frac{1}{2\sqrt{2}\pi N\zeta}\sqrt{\sqrt{\epsilon'^2 + \epsilon''^2} - \epsilon'}. \tag{7}$$

Equation (6) indicates that the photoionization cross-sections $\sigma(\omega)$ of the atom/molecule in a real system can be obtained by transforming the photoionization cross-sections σ_i of the same particle in an ideal gas using a transformation function $f(N, \epsilon', \epsilon''; \zeta)$. Since $f(N, \epsilon', \epsilon''; \zeta)$ is a function of the number density N and the dielectric constant (ϵ', ϵ'') of the system, it may be referred to as Dielectric Influence Function (DIF). For a dilute and ideal gas, the number density N is so small that there are almost no effective interactions among particles, and DIF$\rightarrow$1 and Eq.(1) becomes the accurate expression for photoionization cross-sections. However, as the number density of the system increases, the influence of the dielectric property on cross-sections becomes more and more important, and the interactions among particles are not negligible for a high-density gas, or liquid, or solid. Under the circumstances the DIF is not close to unity in some energy range, and it includes both the higher power dielectric terms and the interactions among particles. These contributions are not included in Eq.(1). It is noted that the Lax's expression in Eq.(2) is similar to our Eq.(6). The factors in the brackets in Eq.(2) play a similar role to that the DIF factor in Eq.(6) plays. It is seen that the present Equations (3), (4) and (6) are dependent only on the system properties explicitly while Eq.(2) is dependent not only on the system properties (n, ϵ) but also on the electric field intensity E of the external electrical field.

To evaluate the photoionization cross-sections of a condensed system using Eq.(3), or Eq.(4) or Eq.(6), one may measure the dielectric constant ϵ of the material in the case of a given number density N. When the values of the (ϵ', ϵ'') are not available, one may use Eq.(6) to evaluate the $\sigma(\omega)$ of the condensed phase by transforming the cross-sections $\sigma_i(\omega)$ of ideal gas us-

ing the DIF in Eq.(7) and the theoretical values of (ϵ', ϵ''). Equation (6) is also very important to understanding the effects of the dielectric property and the particle interactions in the photoionization processes of the condensed system. The dielectric constant (ϵ', ϵ'') may be obtained as follows. For a cubic crystal system, the well-known Clausius–Mossotti (CM) model[18] for the polarized particles in the system may be used to bridge the microscopic polarizability $\alpha(\omega)[= \eta(\omega)+\mathrm{i}\zeta(\omega)]$ of a polarized particle and the macroscopic dielectric function $\epsilon(\omega)[= \epsilon'(\omega) + \mathrm{i}\epsilon''(\omega)]$ of the N-particle system as[16]

$$\epsilon' = \frac{(3 + 8\pi N\eta)(3 - 4\pi N\eta) - 32(\pi N\zeta)^2}{(3 - 4\pi N\eta)^2 + (4\pi N\zeta)^2}, \quad (8)$$

$$\epsilon'' = \frac{36\pi N\zeta}{(3 - 4\pi N\eta)^2 + (4\pi N\zeta)^2}. \quad (9)$$

Equations (8) and (9) represent the massive effects of N polarized particles which have the same microscopic polarizability (η, ζ) on the dielectric constant (ϵ', ϵ'') of the cubic microsystems. The number density N is of the order of 10^{-6}, $4\pi N$ is of the order of 10^{-5} for most ideal gases; while N and $4\pi N$ are of the order of 10^{-4} and 10^{-3} respectively for most real gases; and N and $4\pi N$ are of the order of 10^{-3} and 10^{-2} respectively for most cubic crystals. The DIF $f(N, \epsilon', \epsilon''; \zeta)$ in Eq.(7) may have different forms for photoionization systems in different physical phases or models. When the CM model is used for a cubic crystal, or liquid system, or real gas, $f(N, \epsilon', \epsilon''; \zeta)$ would have the following form:[16]

$$\begin{aligned} f_{\mathrm{CM}}(N, \epsilon', \epsilon''; \zeta) =& \frac{1}{2\sqrt{2}\pi N\zeta}\sqrt{\sqrt{\epsilon'^2 + \epsilon''^2} - \epsilon'} \\ =& \frac{9}{(3 - 4\pi N\eta)^2 + (4\pi N\zeta)^2}\frac{1}{\sqrt{\epsilon'}}\left[1 + \sum_{m=2}^{\infty}(-1)^{m+1}\frac{2(2m-3)!!}{(2m)!!}\left(\frac{\epsilon''^2}{\epsilon'^2}\right)^{m-1}\right]^{1/2}. \end{aligned} \quad (10)$$

This $f_{\mathrm{CM}}(N, \epsilon', \epsilon''; \zeta)$ contains both particle interactions and all higher power terms in the expansion of $\epsilon^{1/2}$. Therefore, the photoionization cross-sections of a cubic solid or real gas system can be obtained through Eq.(6) by transforming the ideal gas cross-sections σ_i in Eq.(1) using the DIF in Eq.(7) or Eq.(10).

The complex dielectric constant (ϵ', ϵ'') in Eq.(7) or Eq.(10) is a function of the number density N of particles of the cubic system and the particle dipole polarizability (η, ζ) in the CM model expressed as in Eqs.(8) and (9). The imaginary part ζ of the dipole polarizability α can be obtained from Eq.(1) using the single particle quantum quantity, i.e. the photoionization cross-sections $\sigma_i(\omega)$ of an ideal gas. The real part η of the dipole polarizability can then be determined by using the imaginary part ζ and the Kramers–Kronig (KK) mathematical transformation relation.[20]

3. Applications

The photoionization cross-sections and the DIF for atomic silver in the cases of different number densities are studied using Eq.(6) and Eq.(7). The experimental values[21] of the photoionization cross-sections $\sigma_i(\omega)$ of a silver ideal gas are used to generate the imaginary part ζ of the silver dipole polarizability $\alpha(\omega)$ from Eq.(1). The real part η of the $\alpha(\omega)$, and the complex dielectric constant (ϵ', ϵ'') of the silver in a cubic crystal are obtained using the KK transformation and Eqs.(8) and (9) respectively.

Figures 1 and 2 present the variations of the real part ϵ' and the imaginary part ϵ'' of the dielectric constant ϵ with both the photon energy ω(eV) and the number density N respectively. Both ϵ' and ϵ'' have structures in a high-density and lower energy region. It is noted that ϵ' has some negative values in a small area, while ϵ'' is always positive. Figure 3 shows the variation of the DIF in Eq.(7) with (ω, N). The DIF has a similar shape and structure to those the real part ϵ' of the dielectric constant ϵ has, but the DIF is always positive while the ϵ' has negative values in a high density and low energy (the most lower left) region in Fig.1. It is seen that the DIF is almost unity in a low-density region where the system is the ideal or dilute gas. This fact indicates that the DIF has no effect on the photoionization cross-sections of an ideal gas. The peak of the structure of the DIF (at about 100eV) is getting sharper as the number

density increases. Therefore, the dielectric property and the particle interactions play a more and more important role in the photoionization process as the number density increases, and their influences on the photoionization cross-sections of a high-density system can be measured by the DIF. Figure 4 presents the variations of the photoionization cross sections of atomic silver with both photon energy ω and particle density N. It is noted that at photon energies above 125eV, the basic shape of the cross-sections in higher densities is similar to that in lower densities, and at energies above 140eV, the cross sections of solid silver are almost identical to those of the silver ideal gas. It is also seen from Figs.1, 3 and 4 that at the density $N' = 1.69 \times 10^{-3} a_0^{-3}$, the real part ϵ', the DIF and the cross-sections of silver show a similar character in structure. In a region for $N < N'$, the physical quantities may be seen as those of the ideal gas where the number density effects have less influence on the photoionization cross-sections, while for $N > N'$, they are the quantities of high-density system where the photoionization cross-sections have a notable change. Therefore, N' may be seen as the Density Turning Point (DTP) of a photoionized system. The DTP is a critical density point at which the photoionization properties of the high-density system start to have notable differences from those of ideal gas for the same atom.

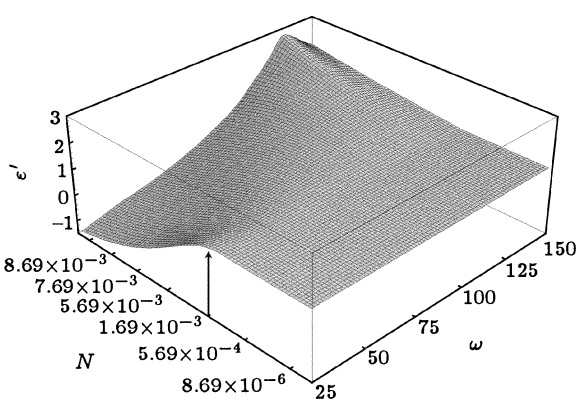


Fig.1. The real part ϵ' of dielectric constant ϵ of atomic silver vs photon energy ω in the cases of different number densities N.

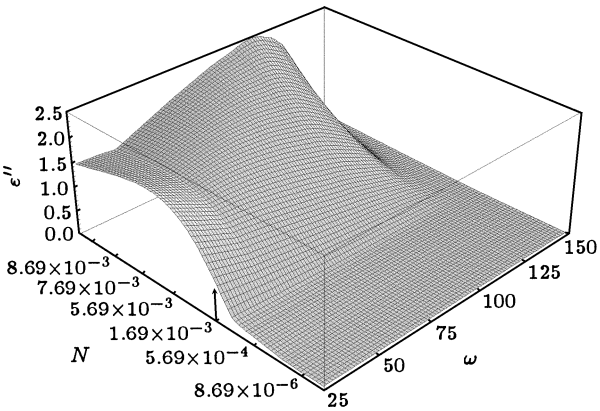


Fig.2. The imaginary part ϵ'' of dielectric constant ϵ of atomic silver vs photon energy ω in the cases of different number densities N.

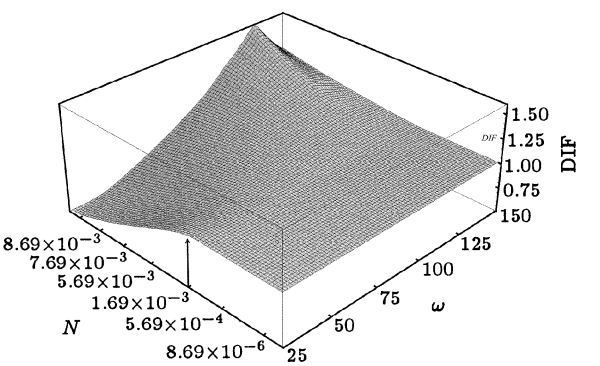


Fig.3. The dielectric influence function DIF $f_{\rm CM}(N, \epsilon', \epsilon''; \zeta)$ of atomic silver vs photon energy ω in the cases of different number densities N.

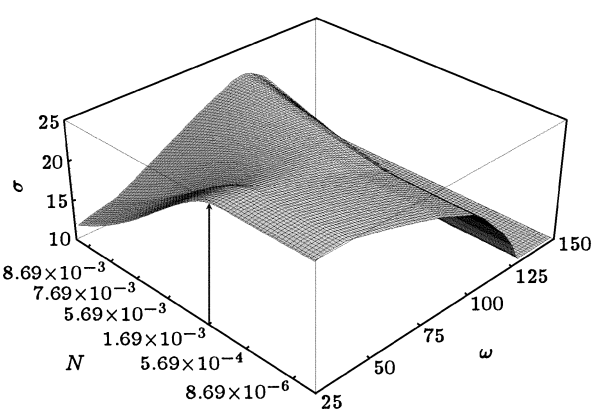


Fig.4. The photoionization cross-sections $\sigma(N, \omega)$ of atomic silver vs photon energy ω in the cases of different number densities N.

Figure 5 shows the comparison of the silver photoionization cross-sections among the solid phase experiments ("○"),[22] determined by using synchrotron radiation on thin films, the ideal gas experimental results σ_i ("– – –")[21] obtained from the photoabsorption experiments, and the present theoretical results ("—") calculated with the solid phase number density $N = 8.69 \times 10^{-3} a_0^{-3}$ at room temperature. It should be mentioned that the theoretical cross sections of solid silver are scaled by a constant[23,24] to the experimental cross sections of solid silver. This shift originates from the fact that the binding energy of the ideal gas differs from that of the solid phase of atomic silver. The present theoretical photoionization cross-sections for solid silver are generated by transforming the ideal gas photoionization cross-sections σ_i using Eq.(6) and the DIF in Eq.(7) where the complex dielectric constant (ϵ', ϵ'') is evaluated using Eqs.(8) and (9) based on the widely used Clausius–Mossotti model for a cubic crystal. In Fig.5, the inset shows the structure of DIF below 200eV, and describes atomic interactions in the photoionized solid silver. For a solid silver system, the general behaviour of the theoretical results ("—") is in good agreement with that of the experimental cross-sections, particularly in a high-energy region. The present example shows that the general expression in Eq.(6) is a useful mean to generate photoionization cross-sections of a high density system by transforming those of ideal gas using a DIF function, and that the differences in cross-section of the same particles between solid phase and ideal gas are mainly caused by the interactions among particles. The differences between present theoretical results ("—") and the experimental photoionization cross-sections ("○") for solid silver at energies below 85eV in Fig.5 might be partly caused by the errors embedded in the classical CM model and the fact that some important quantum effects of solid phase are not included in the model.

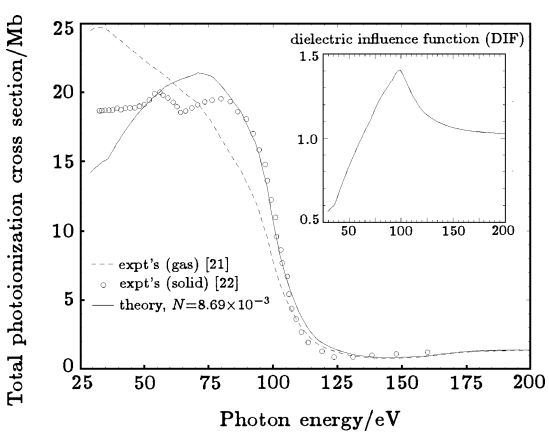


Fig.5. Total photoionization cross-sections of silver atom: "○", the solid silver experimental data; "– – –", the ideal gas experimental results; "—", the present theoretical studies of solid silver. The curve in the inset is the dielectric influence function of the photoionization cross sections of solid silver.

4. Conclusion and discussion

The general expressions for photoionization cross-sections in Eqs.(3) and (6) proposed recently[16] have been used here to study the photoionization cross-sections of solid silver. The new expressions are only based on the experimental Beer–Lambert's law without using any mathematical approximation and physical model, and are the correct functions of macroscopic properties such as the particle number density N and the complex dielectric constant (ϵ', ϵ'') which implicitly measures the particle interactions of the high density photoionized system and the polarization effects of surrounding (environmental) polarized particles on the photoionized atom/molecule. Equation(6) indicates that the photoabsorption (photoionization) cross-sections of atoms in a real system can be obtained by transforming the cross-sections of the same atoms in an ideal gas using the DIF, and that the particle interactions and the polarization effects of the high density photoionized system can be measured by the DIF. If one has the complex dielectric constant (ϵ', ϵ'') of a high density system, the photoionization cross-section of the system can also be directly obtained from Eq.(3). However, Eq.(3) does not give a measure of the particle interactions and the environmental polarization effects of the high density photoionization system.

The energy- and density-dependent photoionization cross-sections of the silver atoms in a solid phase show that density effects are quite important in photoionization studies of a high-density system. A DTP factor is suggested as the critical density point beyond which the photoionization properties of a high-density system notably differs from those of its ideal gas. The present theoretical cross-sections of solid silver are in good agreement with the measured data ("○") in both the shape and the magnitude. The differences of the solid results between experiment and theory at low photon energies may partly originate from experimental errors such as those in the deconvolution procedure and from theoretical approximations used in the CM model. They may also partly come from the errors

in the experimental cross-sections of ideal gas at low energies in Fig.5, where the experimental results of ideal gas failed to show a similar structure to that in the experiments of solid silver at around 65eV. For example, the experimental cross-sections of the barium atom in both solid phase and ideal gas show very similar physical structures.[16] To improve the accuracy of theoretical photoionization cross-sections of a solid system, one may develop a more general theory used to evaluate the dielectric constant which is a function of the particle polarizability $\alpha(\omega)$, and does not depend on a particular physical model like the CM model. One may directly use the measured complex dielectric constants to calculate the photoabsorption (photoionization) cross-sections using Eq.(3). But one should be careful in determining the number density at which the dielectric constants are measured.

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