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Optical absorption of transition-metal island films: Correlation interaction between conduction electrons of transition metals

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Optical plasma-resonance absorption of Pt island films consisting of Pt particles larger than about 25 Å in diameter has been measured in the photon energy range of 0.5–6.5 eV. As in Rh and Pd island films reported previously, the broadening of the optical plasma-resonance absorption reflects a correlation interaction between conduction electrons. Comparison of the broadening for the Pt island films with that for the Rh island films shows that the correlation interaction is strong when the conduction-electron density n is low. In an electron-gas model, the correlation interaction between electrons becomes stronger with lowering electron density, because the magnitude ratio of the Coulomb to kinetic energy increases as the electron density lowers. Thus, the strong correlation-interaction at low n proves that the correlation interaction in transition metals becomes stronger with magnitude ratio. The magnitude ratio in transition metals is pointed out to increase with lowering n and/or with strengthening d character of conduction electrons. Based on the correlation interaction, reflected by the broadening for the Pt, Rh, and Pd island films, and on the strong correlation interaction, found previously for Ir, the order of magnitude ratio is Ir>Pt>Pd>Rh. © *2000 American Institute of Physics.* [S0021-8979(00)05219-1]

I. INTRODUCTION

An electron-gas model, useful as an approximation to metals, is helpful for the study of the correlation interaction between electrons.[1] In the model, the correlation interaction becomes stronger with the magnitude ratio of the Coulomb to kinetic energy of the electron system. This magnitude ratio increases when the electron density lowers, so that the correlation interaction becomes stronger with lowering electron density.

The electron-density dependence of the correlation interaction is informative in the study of a correlation interaction between conduction electrons of metals. For example, when the correlation interaction in the metals is found to be strong at the low conduction-electron density, we see that the correlation interaction becomes stronger with the magnitude ratio.

In transition metals, interactions between electrons significantly influence electronic properties.[2] Many studies of these have been reported,[3] but few of the interactions have been well understood.

Metal island films consisting of small metal particles show optical plasma-resonance absorption arising from plasma oscillations of conduction electrons in the small metal particles.[4,5] The plasma-oscillation damping, which reflects the motion of the conduction electrons, results in the broadening of the optical plasma-resonance absorption.[4,5] Thus, the broadening reflects factors, such as interactions between conduction electrons, in the motion.

A previous study[6] of the optical plasma-resonance absorption of Ni and Pd island films has shown that a correlation interaction, based on d character of conduction electrons, occurs between the conduction electrons, and that the correlation interaction causes the plasma-oscillation damping and is reflected by the broadening of the optical plasma-resonance absorption. In the study,[6] as in those before,[5] the broadening was measured as the half width of the optical plasma-resonance absorption.

From comparison of the half width (i.e., the broadening) for Ni, Pd, fcc Co, and Rh island films, the correlation interaction has been studied further. In fcc Co and Ni island films,[7] the correlation interaction was reported to become stronger with strengthening d character. In Rh and Pd island films,[8] the correlation interaction in Pd was found to be stronger than that in Rh, although the d character is weaker in Pd than in Rh. From this discrepancy and the low conduction-electron density of Pd, it was suggested that the correlation interaction is strong also when the conduction-electron density n is low.[8]

Because the d character contributes to the correlation interaction, comparison of the half width for island films of transition metals, in which the d character is comparable and n is different, should provide reliable data on the correlation interaction at low n. Therefore, the earlier suggestion based on Rh and Pd, which have the different d character and n, is insufficient as evidence that the correlation interaction is strong at low n.

In this study, the half width for Pt and Rh[8] island films is compared. The d character in Pt is comparable to that in Rh, and the n of Pt is about one-third that of Rh. The comparison shows the correlation interaction to be strong at low n, which proves that the correlation interaction becomes stronger with magnitude ratio of the Coulomb to kinetic energy. This ratio is pointed out to increase with lowering n and/or with strengthening d character. For Pt, Pd, Rh, and Ir, the order of the magnitude ratio is discussed based on the correlation

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interaction, reflected by the broadening (i.e., by the half width) for the Pt, Pd,[6] and Rh[8] island films, and on the strong correlation interaction in Ir,[9] which results in the conduction-electron localization.

II. EXPERIMENT

The experiment was carried out as previously.[6–9] In a vacuum chamber, electron-microscopic meshes covered with a carbon film and a fused-quartz substrate ($18\times18\times0.5\,mm^3$) were placed above an evaporation source. The meshes and substrate were adjacent, and the distance from the evaporation source was the same (30.3 cm) for the meshes and substrate.

In an oil-free vacuum of $\sim10^{-8}$ Torr, by electron beam heating, SiO_2 was first deposited both on the meshes and on the fused-quartz substrate. Next, at pressures of $\sim10^{-7}$ Torr,Pt (purity 99.9%) was deposited in order to obtain island films. The films were then annealed for 1 h. During deposition and annealing, the meshes and substrate were held at about 500 °C. After annealing, the films were coated with SiO_2 (thickness about 300 Å) to prevent adsorption or chemical reactions on exposure to air and then cooled to room temperature at a rate of about 1–2 °C/min. The transmittance of the evaporated SiO_2 film without Pt particles was almost constant within the spectral range of interest here.

Optical and electron-microscopic investigation were carried out after exposure of the samples to air. In the photon energy range of 0.5–6.5 eV, transmittance spectra for normal incidence and their derivatives for a wavelength difference of about 4 nm were measured within experimental accuracy of ±0.1% and ±(0.001–0.01) eV at room temperature and −193 °C with a double-beam spectrophotometer. The particle size and the electron-diffraction pattern were investigated with an electron microscope operating at 200 kV.

III. RESULTS AND DISCUSSION

In Sec. III A, optical plasma-resonance absorption of Pt island films is identified, and the broadening of the optical plasma-resonance absorption is shown to result from a correlation interaction. In Sec. III B, the dependence of the correlation interaction in transition metals on the magnitude ratio of the Coulomb to kinetic energy is analyzed based on comparison of the broadening of the optical plasma-resonance absorption of transition-metal island films.

A. Optical plasma-resonance absorption and its broadening of Pt island films

An example of the transmittance spectrum and its derivative of Pt island films at room temperature is shown in Fig. 1(a), in which only broad absorption, not found in bulk Pt,[10] appears at about 3.7 eV.

Figure 1(b) shows the electron micrograph of the film of Fig. 1(a). The contrast of the particles in this micrograph and in the following electron micrographs is not uniform because of the diffraction contrast, showing the particles in this study to be polycrystalline.[11]

The particle-size distribution in volume percent of the film of Fig. 1(a) is shown in Fig. 1(c). As shown in this figure, two peaks appeared in the particle-size distribution when the weight thickness is larger than about 20 Å.

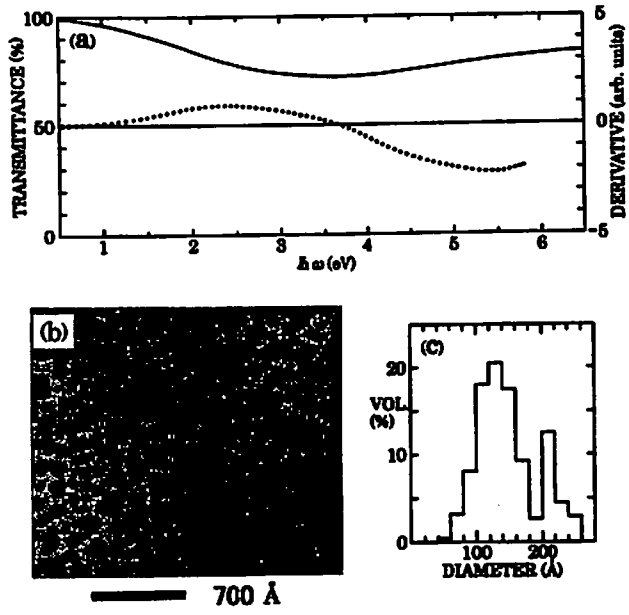

FIG. 1. (a) Transmittance spectrum (solid curve) and its derivative (dotted curve) of a Pt island film. The weight thickness is 22.8 Å. The deposition rate was 0.06 Å/s. The half width $2\Gamma_{1/2}$ of the optical plasma-resonance absorption is 3.35 eV. (b) Electron micrograph of the same island film. (c) Particle-size distribution in volume percent of the same island film.

Figures 2–5 show the transmittance spectrum, its derivative, the electron micrograph, and the particle-size distribution in volume percent of the Pt island film at room temperature with particle diameters of about 110, 70, 45, and 25 Å, respectively.

In Figs. 1(a)–5(a), the broad absorption shifts to higher energies with decreasing weight thickness. Such a shift is characteristic of the optical plasma-resonance absorption of metal island films,[12–14] and thus shows the broad absorption to be the optical plasma-resonance absorption.

In this study, evaluation of the half width of the optical plasma-resonance absorption is based on the formula for the transmittance of continuous-thin metal films. We consider here a continuous-thin metal film of the dielectric constant ε, which is on the transparent substrate of the refractive index n_s, and regard the film thickness d to be smaller than the wavelength λ of light ($2\pi d \ll \lambda$). In this case, when the film, the substrate, and the medium of the refractive index n_i, from which light is incident, are optically isotropic, the transmittance T for normal incidence of light is approximated as[15]

$$T/T_s=\left[1+\frac{2}{n_i+n_s}\frac{2\pi}{\lambda}d\,\mathrm{Im}(\varepsilon)\right]^{-1}.$$

Here $T_s[=4n_in_s/(n_i+n_s)^2]$ is the transmittance for the bare surface of the substrate and $\mathrm{Im}(\varepsilon)$ is the imaginary part of ε. The ratio T/T_s is measured directly when a double-beam spectrophotometer is used with a bare substrate as a reference sample. The second term of the right side gives the absorption spectrum.

The earlier formula for T/T_s is applicable to experimental transmittance spectra of metal island films, the weight thickness d_w of which is smaller than the wavelength λ of

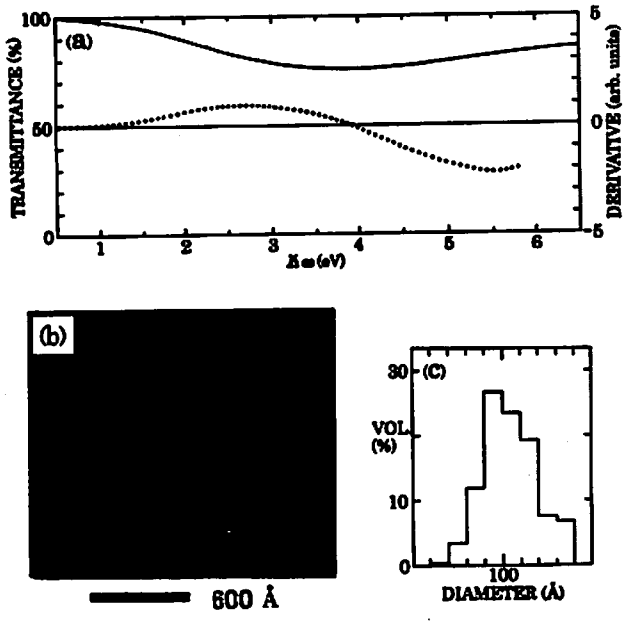

FIG. 2. (a) Transmittance spectrum (solid curve) and its derivative (dotted curve) of a Pt island film with a particle diameter of about 110 Å. The weight thickness is 19.0 Å. The deposition rate was 0.06 Å/s. The half width $2\Gamma_{1/2}$ of the optical plasma-resonance absorption is 3.35 eV. (b) Electron micrograph of the same island film. (c) Particle-size distribution in volume percent of the same island film.

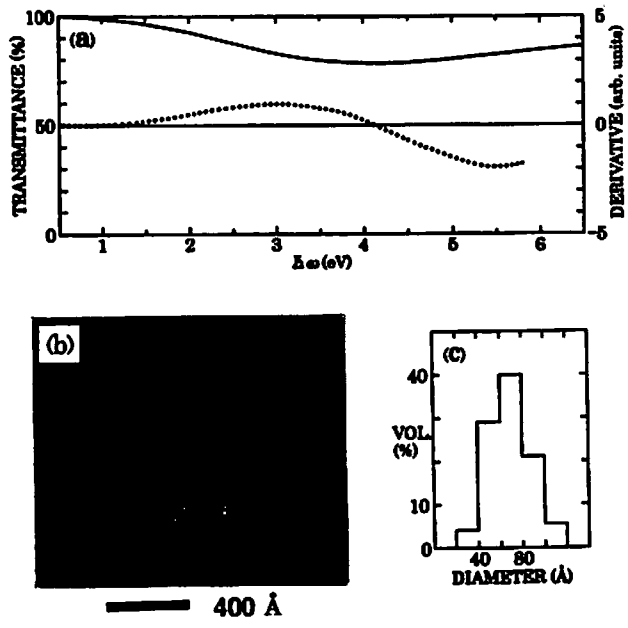

FIG. 3. (a) Transmittance spectrum (solid curve) and its derivative (dotted curve) of a Pt island film with a particle diameter of about 70 Å. The weight thickness is 15.2 Å. The deposition rate was 0.06 Å/s. The half width $2\Gamma_{1/2}$ of the optical plasma-resonance absorption is 3.35 eV. (b) Electron micrograph of the same island film. (c) Particle-size distribution in volume percent of the same island film.

light ($2\pi d_w \ll \lambda$).[16–18] As previously described,[6–8] the formula was applied to the experimental transmittance spectra of the Pt island films in this study, and the half width of the optical plasma-resonance absorption was obtained from $\hbar\omega$ [$=1239\,(\text{eV nm})/\lambda(\text{nm})$] at which absorption is half of the peak of the optical plasma-resonance absorption.

The earlier $\hbar\omega$ at the high-energy flank was out of the spectral range of interest here. Thus, as before,[6–8] the half width was defined in terms of $2\Gamma_{1/2}$, where $\Gamma_{1/2}$ is half of the half width at the low-energy flank. The $2\Gamma_{1/2}$ for the Pt island films was almost constant (3.35–3.40 eV) as shown in Figs. 1(a)–5(a).

In transition metals, since the Fermi level falls in d bands and s and d bands are hybridized (s–d hybridization),[2,19,20] d character of conduction electrons is strong. In the previous study of Ni and Pd island films,[6] it was shown that, in the dynamical state due to the incidence of light, a correlation interaction acts between conduction electrons because of a localizing tendency due to the d character, and that the correlation interaction, which causes the plasma-oscillation damping, is reflected by the broadening of the optical plasma-resonance absorption. The broadening was expressed in terms of the half width ($2\Gamma_{1/2}$) of the optical plasma-resonance absorption, as in previous studies.[5]

The previous study[7] of fcc Co and Ni island films reported that the $2\Gamma_{1/2}$ for the fcc Co island films is larger than that for the Ni island films. From this, the correlation interaction, reflected by the broadening (i.e., by $2\Gamma_{1/2}$), has been seen to become stronger with strengthening d character, because fcc Co has stronger d character than Ni.[7]

It was reported that the optical plasma-resonance absorption is absent in Ir (Ref. 9) and Fe and Cr (Ref. 21) island films. This absence was attributed to the conduction-electron localization; i.e., a strong correlation interaction, due to the strong d character based on the large s–d hybridization (Ir)[9] or on the Fermi level deeply falling in d bands (Fe and Cr),[21] causes the localization. This supports the earlier d-character dependent strengthening of the correlation interaction.

In the previous studies,[6,7] the following facts have been shown as evidence of the broadening reflecting the correlation interaction: $2\Gamma_{1/2}$ is almost constant irrespective of the variety of particle shapes and the particle-size distribution; and the optical plasma-resonance absorption is independent of temperature.

The almost constant $2\Gamma_{1/2}$ was derived from agreement between $2\Gamma_{1/2}$ of experimental optical plasma-resonance absorption and $2\Gamma_{1/2}$ of simulated optical plasma-reonance absorption based on the earlier formula for T/T_s and bulk optical constants.[6] The temperature independence of the optical plasma-reonance absorption is because the correlation interaction in the optical plasma-reonance absorption is the Coulomb interaction, which does not depend on temperature, between conduction electrons of antiparallel spin.[6]

The $2\Gamma_{1/2}$ for the Pt island films is almost constant [3.35–3.40 eV, Figs. 1(a)–5(a)] irrespective of the variety of particle shapes [Figs. 1(b)–5(b)] and the particle-size distribution [Figs. 1(c)–5(c)], as shown earlier. The optical plasma-reonance absorption of the Pt island films did not change as the temperature was decreased from room temperature to −193 °C, which shows the optical plasma-reonance absorption to be temperature independent. These results agree with the earlier evidence, showing that the broadening of the optical plasma-reonance absorption of the Pt island films reflects the correlation interaction.

For the optical plasma-reonance absorption of metal is-

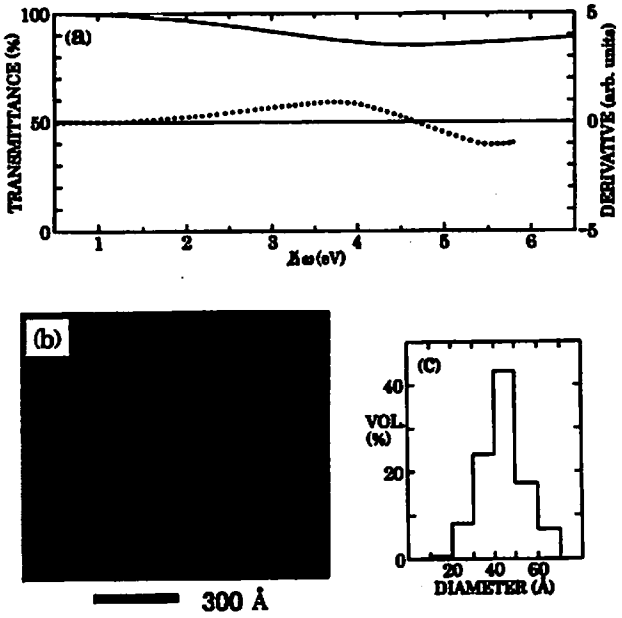

FIG. 4. (a) Transmittance spectrum (solid curve) and its derivative (dotted curve) for a Pt island film with a particle diameter of about 45 Å. The weight thickness is 9.5 Å. The deposition rate was 0.06 Å/s. The half width $2\Gamma_{1/2}$ of the optical plasma-resonance absorption is 3.40 eV. (b) Electron micrograph of the same island film. (c) Particle-size distribution in volume percent of the same island film.

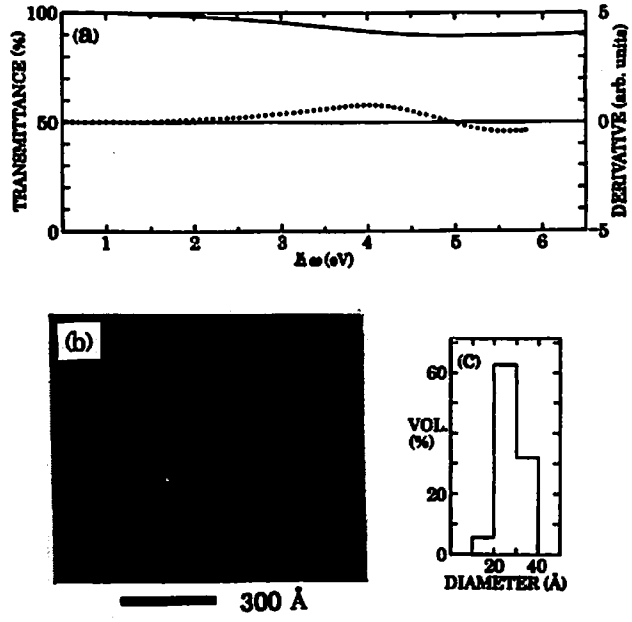

FIG. 5. (a) Transmittance spectrum (solid curve) and its derivative (dotted curve) of a Pt island film with a particle diameter of about 25 Å. The weight thickness is 5.7 Å. The deposition rate was 0.05 Å/s. The half width $2\Gamma_{1/2}$ of the optical plasma-resonance absorption is 3.36 eV. (b) Electron micrograph of the same island film. (c) Particle-size distribution in volume percent of the same island film.

land films, it has been reported theoretically that the disorder in the particle distribution contributes to the broadening of the optical plasma-reonance absorption.[22] In the previous study,[6] $2\Gamma_{1/2}$ of experimental optical plasma-reonance absorption agreed well with $2\Gamma_{1/2}$ of simulated optical plasma-reonance absorption based on the earlier formula for T/T_s and bulk optical constants. The $2\Gamma_{1/2}$ of the simulated optical plasma-reonance absorption is due to only the damping of the plasma oscillation and has no relation to the disorder.[6] Thus, the disorder is considered to have little effect on the almost constant $2\Gamma_{1/2}$ of experimental optical plasma-reonance absorption of transition-metal island films.

There seems to be very little data on interactions between Pt and SiO_2. In this study, therefore, interactions between the Pt particles and the SiO_2 matrix were not taken into consideration.

In this study, only the fcc structure could always be identified in electron diffraction patterns of the Pt island films. From this result, chemical reactions such as oxidation are considered to occur rarely, and so the formation of a compound layer (such as oxide layer) on the surface of the Pt particles was not taken into account.

In bulk metals, absorption due to conduction electrons conspicuously appears in the low energy-range below about 1.5 eV.[23,24] The presence of the absorption makes it difficult to investigate the existence of interband absorption in the low energy range. Because such absorption does not occur in metal island films, the optical study of the metal island films gives reliable data on the existence of the interband absorption in the low energy range.[9]

A study[10] of optical properties of bulk Pt has reported the existence of interband absorption at about 0.7 eV. The interband absorption is not found in Figs. 1(a)–5(a). As the optical study of metal island films is reliable, this absence in Figs. 1(a)–5(a) indicates that the interband absorption at about 0.7 eV, reported previously, does not exist.

As shown in the previous study,[6] comparison of experimental optical plasma-reonance absorption with simulated optical plasma-reonance absorption based on the earlier formula for T/T_s and bulk optical constant is useful for the study of the correlation interaction. For bulk Pt, there seems to be very little data on optical constant, which is corrected for the nonexistence of the interband absorption at about 0.7 eV. For this reason, the simulation of the optical plasma-reonance absorption was difficult in this study.

B. Dependence of the correlation interaction on the magnitude ratio of the Coulomb to kinetic energy

As mentioned in Sec. I, by finding the correlation interaction for metals to be strong at the low conduction-electron density, we can prove that the correlation interaction becomes stronger with magnitude ratio of the Coulomb to kinetic energy.

It has been shown in the previous studies that the correlation interaction appears in the optical plasma-reonance absorption and is reflected by the broadening of the optical plasma-reonance absorption.[6,7] Here, as previously described,[6,7] the broadening is expressed in terms of $2\Gamma_{1/2}$, and the correlation interaction at the low conduction-electron density is discussed based on comparison of $2\Gamma_{1/2}$.

Taking into account the contribution of the d character to the correlation interaction, reliable data on the correlation interaction at the low conduction-electron density should be obtained from comparison of $2\Gamma_{1/2}$ for island films of tran-

sition metals, which satisfy the condition that the d character is comparable and the conduction-electron density n is different. Here, Pt and Rh are selected as such metals.

In the previous study of the correlation interaction for Rh and Pd island films,[8] the earlier condition was not satisfied (i.e., Rh and Pd have different d character and n), so although it was suggested that the correlation interaction is strong at low n, it could not be concluded.

Here, first the d character of conduction electrons and the n of Pt and Rh are discussed. Then, the correlation interaction at low n is investigated based on comparison of the $2\Gamma_{1/2}$ for the Pt island films with that for the Rh island films reported previously.[8]

Factors for d character of conduction electrons of transition metals are the position of the Fermi level relative to d bands and the $s-d$ hybridization: d character is strong when the Fermi level is positioned deeply in d bands and/or when the $s-d$ hybridization is large.

There has been very little quantitative data on d character based on the position of the Fermi level relative to d bands. In Pt and Rh, which have the fcc structure and are paramagnetic, the Fermi level intersects the upper part of d bands.[19,20] In energy band structures,[19] the position of the Fermi level relative to d bands is shallower in Pt than in Rh. However, the difference in the position is small. For this reason, the difference in the d character based on the position of the Fermi level between Pt and Rh is also presumably small.

There has been only a small amount of data on the $s-d$ hybridization of transition metals. Here, we refer to the $s-d$ hybridization coefficient B used in the calculation of energy bands by Smith[19] (Table I, in which the B of Pd is also shown). In Table I, the B of Rh is about 97% of that of Pt; that is, the B of Pt is nearly equal to that of Rh. Thus, the $s-d$ hybridization of Pt can be regarded to be equal to that of Rh, implying that there is very little difference in d character based on the $s-d$ hybridization between Pt and Rh.

From the earlier discussion of the d character, it is reasonable to consider that the d character of Pt is comparable to that of Rh.

The n of Pt and Rh was calculated based on the theoretical values of the number of conduction electrons per atom (Pt:0.4055; Rh:1.0989)[25,26] and on the lattice constant (Pt: 3.9231 Å; Rh:3.8043 Å),[27] as previously described.[8] The calculated n is also shown in Table I, in which the n of Pd is also shown (the number of conduction electrons per atom and the lattice constant of Pd are, respectively, 0.3734 and 3.8898 Å).[25,27] As shown in Table I, the n of Pt is about 34% of that of Rh. That is, Pt has very low n compared to Rh.

The $2\Gamma_{1/2}$ for the Pt and Rh[8] island films is shown in Table II, in which the $2\Gamma_{1/2}$ for Pd island films reported previously[6] is also shown. The $2\Gamma_{1/2}$ for the Pt island films is large compared to that for the Rh island films. This shows that the correlation interaction is strong at low n, and thus proves that the correlation interaction in transition metals becomes stronger with magnitude ratio of the Coulomb to kinetic energy.

The contribution of the d character to the correlation interaction is confirmed below. Since n contributes to the correlation interaction as shown earlier, the contribution of the d character should be confirmed for transition metals, in which n is almost equal and the d character is different. Pt and Pd are selected as such metals.

TABLE I. The $s-d$ hybridization coefficient B and the conduction-electron density n of Pt, Rh, and Pd.

	Pt	Rh	Pd
B (Ry)[a]	1.76	1.70	1.42
$n(10^{22}/cm^3)$[b]	2.69	7.98	2.54

[a]Data from Ref. 19, in which B is in Rydberg.
[b]Calculation based on the number of conduction electrons per atom in Ref. 25 and on the lattice constant in Ref. 27.

Pd has the fcc structure and is paramagnetic. In Table I, the n of Pd is about 94% of that of Pt, i.e., the difference in n between Pd and Pt is very small. Therefore, the n of Pd can be regarded as equal to that of Pt.

In Pd, the Fermi level intersects the upper part of d bands as in Pt.[19,20] The position of the Fermi level relative to d bands is deeper in Pt than in Pd, but the difference in the position is small.[19] Similar to the earlier case of Pt and Rh, the difference in the d character based on the position of the Fermi level between Pt and Pd is also presumably small. The $s-d$ hybridization of Pt is larger than that of Pd: the B of Pt is about 24% larger than that of Pd (Table I). Thus, the d character based on the $s-d$ hybridization is stronger in Pt than in Pd. From this discussion of the d character, it is reasonable to consider that Pt has stronger d character than Pd.

In Table II, the $2\Gamma_{1/2}$ for the Pt island films is larger than that for the Pd island films. This confirms that the correlation interaction becomes stronger with strengthening d character. From this confirmation and the dependence of the correlation interaction on the magnitude ratio, we see that the magnitude ratio in transition metals increases with strengthening d character.

As mentioned in Sec. III A, the previous study[6] showed that the d character has a significant effect on the correlation interaction. That is, the d character causes conduction electrons to have a localizing tendency, and based on this tendency, the motion of the conduction electrons is correlated strongly in the dynamical state due the incidence of light (i.e., a correlation interaction acts between the conduction electrons). From this, it is seen that the d character influences the motion of the conduction electrons.

Since the earlier motion is related to the Coulomb and kinetic energy of the conduction-electron system, the d character affects the Coulomb and kinetic energy. Presumably, the d character affects also the magnitude ratio of the Coulomb to kinetic energy. In this study, it was difficult to clarify the mechanism of the earlier magnitude-ratio increase due to the strengthening of the d character.

It should be noted that, in transition metals, not only n but also the d character is the factor contributing to the magnitude ratio: The magnitude ratio in transition metals increases with lowering n and/or with strengthening d character.

TABLE II. The half width $2\Gamma_{1/2}$ of the optical plasma-resonance absorption of the Pt, Rh, and Pd island films.

	Pt	Rh	Pd
$2\Gamma_{1/2}$ (eV)	3.35–3.40	2.36–2.40[a]	2.50–2.60[b]

[a]Data from Ref. 8
[b]Data from Ref. 6.

The half width of the optical plasma-resonance absorption corresponds to the broadening of the optical plasma-resonance absorption, and the broadening reflects the correlation interaction, which becomes stronger with magnitude ratio of the Coulomb to kinetic energy. Therefore, the order of $2\Gamma_{1/2}$ should show the order of the magnitude ratio. We can see from the increasing order of $2\Gamma_{1/2}$ in Table II that the magnitude ratio increases from Rh to Pd to Pt.

Ir has the fcc structure and is paramagnetic. In the previous study of a correlation interaction for Ir island films,[9] it was shown that *d* character of conduction electrons of Ir is strong because of the large $s-d$ hybridization, and that a strong correlation interaction due to the strong *d* character leads to the conduction-electron localization.

The correlation interaction, which causes the conduction-electron localization in Ir, is obviously strong compared to the correlation interaction, which appears in the optical plasma-reonance absorption of the Pt island films. Thus, for Pt, Rh, Pd, and Ir, the order of magnitude ratio is Ir>Pt>Pd>Rh.

Island films of 3*d*fcc transition-metals (Ni[6] and fcc Co[7]) also show optical plasma-reonance absorption. These metals are ferromagnetic and thus have energy bands for electrons of majority and minority spins.[28–31] These energy bands differ in the position of the Fermi level relative to *d* bands and the $s-d$ hybridization. It is thus difficult to compare the position of the Fermi level and the $s-d$ hybridization of these metals with those of Pt, Rh, and Pd. In this study, therefore, we did not refer to the half width $2\Gamma_{1/2}$ of the optical plasma-reonance absorption of the 3*d*fcc transition-metal island films.

IV. SUMMARY

In an electron-gas model, a correlation interaction between electrons becomes stronger with magnitude ratio of the Coulomb to kinetic energy. The magnitude ratio increases as the electron density lowers. Thus, the correlation interaction becomes stronger with lowering electron density.

From the electron-density dependence of the correlation interaction, it can be seen that if a correlation interaction between conduction electrons of metals is found to be strong at a low conduction-electron density, the correlation interaction becomes stronger with magnitude ratio.

For transition metals, previous studies of optical plasma-reonance absorption of island films have reported that a correlation interaction, becoming stronger with strengthening *d* character of conduction electrons, occurs between the conduction electrons, and that the broadening of the optical plasma-reonance absorption reflects the correlation interaction, which causes the plasma-oscillation damping.

In this study, as in previous, the broadening of the optical plasma-reonance absorption was expressed in terms of the half width $2\Gamma_{1/2}$ (where $\Gamma_{1/2}$ is half of the half width at the low-energy flank) of the optical plasma-reonance absorption, and the correlation interaction at the low conduction-electron density was investigated based on comparison of $2\Gamma_{1/2}$.

Considering the contribution of the *d* character to the correlation interaction, comparison of $2\Gamma_{1/2}$ for island films of transition metals, having the comparable *d* character and the different conduction-electron density, should give reliable data on the correlation interaction at a low conduction-electron density.

The *d* character in Pt and Rh is comparable, and the conduction-electron density *n* is lower in Pt than in Rh. The $2\Gamma_{1/2}$ for Pt island films was larger than that for Rh island films reported previously, i.e., $2\Gamma_{1/2}$ was large at low *n*. This shows that the correlation interaction is strong at low *n*, and thus proves that the correlation interaction in transition metals becomes stronger with the magnitude ratio.

n is almost equal in Pt and Pd, and Pt has strong *d* character compared to Pd. $2\Gamma_{1/2}$ was larger for the Pt island films than for Pd island films reported previously, i.e., $2\Gamma_{1/2}$ was large at the strong *d* character. This result confirms that the correlation interaction becomes stronger with strengthening *d* character.

From the earlier confirmation and the dependence of the correlation interaction on the magnitude ratio, it is seen that the magnitude ratio in transition metals increases with strengthening *d* character. In this study, the mechanism of this *d*-character-dependent increase in the magnitude ratio could not be clarified.

Note that, in transition metals, the *d* character is the factor for the magnitude ratio as well as *n*: The magnitude ratio in transition metals increases with lowering *n* and/or with strengthening *d* character.

The half width $2\Gamma_{1/2}$ corresponds to the broadening. Thus, the order of $2\Gamma_{1/2}$ should show the order of the magnitude ratio, because the broadening reflects the correlation interaction, which becomes stronger with magnitude ratio. From the $2\Gamma_{1/2}$ for the Pt, Rh, and Pd island films, the magnitude ratio was seen to increase from Rh to Pd to Pt.

It has been shown in the previous study of Ir island films that a strong correlation interaction in Ir, due to the strong *d* character based on the large $s-d$ hybridization, causes the conduction-electron localization.

The correlation interaction resulting in the conduction-electron localization is obviously strong compared to the correlation interaction appearing in the optical plasma-reonance absorption of the Pt island films. Thus, for Pt, Rh, Pd, and Ir, the magnitude ratio is in the order Ir>Pt>Pd>Rh.

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