Plasmon excitation by slow ions

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We present a theoretical approach to the problem of subthreshold plasmon excitation in proton-aluminum collisions. Based both on recent experimental results by Ritzau *et al.* [Phys. Rev. B **59**, 15506 (1999)] and on previous calculations by two of the authors, we solve the master equations and obtain the probability of both plasmon excitation and nearly free electron excitation due to plasmon decay as functions of the projectile initial velocity. The mechanism considered for subthreshold plasmon excitation involves an intermediary fast electron as suggested by Ritzau *et al.*

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I. INTRODUCTION

Plasmon creation is an important energy loss mechanism for fast charged particles travelling inside simple metals. These collective excitations are found theoretically even in simple descriptions of the target metal, such as the jellium model [1], and it is a well-known result that within this model, there is a minimum, or threshold, projectile velocity v_{th} necessary to allow for plasmon excitation. However, recent experimental results [2,3] present evidence of plasmon excitation in proton-aluminum collisions for proton velocities below the threshold.

Different possible explanations for this phenomenon are discussed by Ritzau *et al.* [2] who conclude that the most plausible mechanism for subthreshold plasmon creation is the previous excitation of an intermediary fast enough ($v_e > v_{the}$) valence electron (note that the threshold velocity for a proton projectile v_{thp} is a little lower than the one corresponding to an electron projectile v_{the}). That is, slow protons ($v_i < v_{thp}$) cannot excite plasmons directly but an *indirect* plasmon creation mechanism can take place in which protons excite valence electrons (electron-hole creation) that are fast enough to excite plasmons.

The fact that both Ritzau *et al.* and Lacombe *et al.* work with heavy projectiles forces them, in order to clearly identify plasmon excitation, to analyze the emitted electron spectrum instead of using the energy-loss method, common in studies with fast electrons. Therefore they actually detect electrons excited by plasmon decay and emitted with a characteristic energy signature. Note as well that indirect plasmon excitation cannot be detected by means of the energy-loss method.

Processes that result in plasmon decay occur in real metals due to several damping mechanisms. In particular, low momentum plasmons mostly decay by inducing interband transitions on nearly free electrons [4–6]. This mechanism was thoroughly studied by two of the authors in a series of publications [4,7,8]. In those papers, plasmons were excited *directly* by the fast projectile (either a proton or a primary electron) and decayed transferring their energy and momentum to nearly free electrons.

In the current work, on the other hand, plasmons are generated in slow proton-aluminum collisions ($v_i < v_{thp}$) via intermediary fast electrons (*indirect* mechanism). For this process a theoretical analysis is performed, and expressions for both the probability of plasmon excitation and the probability of nearly free electron excitation due to plasmon decay are obtained by solving the master equations for the number of excited electrons and plasmons in the solid. Results for the *indirect* process are compared to the ones corresponding to the *direct* mechanism, previously obtained in the mentioned articles [4,7,8].

Although Ritzau *et al.* make a very interesting discussion, estimating the emitted electron yield due to plasmon decay contributions, we consider the present work a somewhat more theory-based approach to the subject which complements and completes the results there portrayed.

In Sec. II A, the question of indirect plasmon excitation is addressed. The probability is obtained for a slow proton to excite a plasmon in the electron gas via the excitation of an intermediary fast enough electron.

In Sec. II B, a mechanism that allows for plasmons to decay is added to the previous scenario. The probability is obtained for a slow proton to excite a nearly free electron (NFe) via a three-step process in which the proton excites a fast free electron, this electron excites a plasmon and this plasmon decays exciting a nearly free electron.

In Sec. III, results are presented for the mentioned probabilities, together with the energy spectra for the excited electrons due to plasmon decay. Also, the intermediary-electron energy distribution is displayed.

In Sec. IV, the summary and conclusions of the present work are given. Atomic units are used throughout the paper.

II. THEORY

A. Subthreshold plasmon excitation

The process of interest is that in which a slow proton ($v_i < v_{th}$) excites a plasmon in the electron gas via the intermediary excitation of a fast electron. Schematically,

$$\mathrm{H}_i^+ + e_i \rightarrow \mathrm{H}_f^+ + e_e,$$

$$e_e + \mathrm{FEG} \rightarrow e_f + \mathrm{FEG}^*. \tag{1}$$

The proton flux N_0 is considered constant and the evolution of the number of excited electrons and plasmons is obtained from the master equations

$$\dot{N}_{e,\bar{k}_e}(t) = \alpha(\bar{k}_e)N_0 - \beta^{tot}(k_e)N_{e,\bar{k}_e}(t),$$

$$\dot{N}_{pl,\bar{k}_e}(t) = \beta_2(k_e)N_{e,\bar{k}_e}(t), \tag{2}$$

where $N_{e,\bar{k}_e}(t)$ is the number of electrons in the state $\bar{k}_e$ at time t, $\alpha(\bar{k}_e)$ is the probability per unit time for a proton to excite a valence electron to the state $\bar{k}_e$, $\beta^{tot}(k_e)=\beta_1(k_e)+\beta_2(k_e)$ is the probability per unit time for a valence electron in state $\bar{k}_e$ to excite either another electron (β_1) or a plasmon (β_2) in the jellium, and $N_{pl,\bar{k}_e}(t)$ is the number of plasmons excited by a valence electron in a state $\bar{k}_e$. Note that, as we are inside the bulk and within a jellium model calculation, both β coefficients depend only on $k_e=|\bar{k}_e|$.

The differential equations in Eq. (2) are easily solved and one finds

$$N_{e,\bar{k}_e}(t) = \frac{\alpha(\bar{k}_e)N_0}{\beta^{tot}(k_e)}(1 - e^{-\beta^{tot}(k_e)t}),$$

$$N_{pl,\bar{k}_e}(t) = \frac{\alpha(\bar{k}_e)\beta_2(k_e)N_0}{[\beta^{tot}(k_e)]^2}[e^{-\beta^{tot}(k_e)t} - 1 + \beta^{tot}(k_e)t]. \tag{3}$$

Next, an integration over the intermediary electron states is performed as follows:

$$\begin{aligned} N_{pl}(t) &= N_0 \int \frac{\alpha(k_e)\beta_2(k_e)}{[\beta^{tot}(k_e)]^2}[e^{-\beta^{tot}(k_e)t} - 1 + \beta^{tot}(k_e)t]d\bar{k}_e \\ &= N_0 \int \frac{\beta_2(k_e)}{(\beta^{tot})^2}(e^{-\beta^{tot}t} - 1 + \beta^{tot}t) \\ &\quad \times \underbrace{\int \alpha(\bar{k}_e)\sin\theta_e d\theta_e d\varphi_e k_e^2}_{\alpha(k_e)} dk_e. \end{aligned} \tag{4}$$

Therefore the probability per unit time for a proton to *indirectly* excite a plasmon via the excitation of an intermediary valence electron is given by

$$\begin{aligned} P_t^{Pls} = \frac{N_{pl}(t)}{N_0 t} &= \frac{1}{t}\int \frac{\alpha(k_e)\beta_2(k_e)}{[\beta^{tot}(k_e)]^2} \\ &\quad \times [e^{-\beta^{tot}(k_e)t} - 1 + \beta^{tot}(k_e)t]k_e^2 dk_e. \end{aligned} \tag{5}$$

The probability for indirect plasmon excitation, put forward in Eq. (5), is expressed in terms of the coefficients α, β^{tot}, and β_2 which correspond to probabilities of various direct excitations. The expression for α (probability for a proton to excite a valence electron to states with momentum modulus k_e) was obtained by means of the binary collisional formalism as presented by Arbó and Miraglia [9], which allows one to obtain the probability of a binary excitation as a function of the electron's momentum modulus k_e. On the other hand, the expressions for β_2 (probability for a primary electron with velocity k_e to excite a plasmon in the electron gas) and β^{tot} (probability for a primary electron with velocity k_e to excite either a plasmon or another electron in the electron gas) were taken from the standard dielectric formalism as introduced by Pines [1]. They read

$$\begin{aligned} \alpha(k_e) = \frac{dP_{tp}^{Bin}}{dk_e} &= 2\pi \int \frac{2\delta(\omega - \bar{v}_i \,.\, \bar{p})}{p^4|\epsilon|^2}\left(\frac{4\pi Z_P}{(2\pi)^3}\right)^2 \\ &\quad \times \theta(k_F - k_i)\theta(k_f - k_F)\sin\theta_e d\theta_e d\varphi_e d\bar{k}_i, \end{aligned}$$

$$\begin{aligned} \beta^{tot}(k_e) = P_{te} &= -\frac{2Z_P^2}{\pi k_e}\int_0^{+\infty}\int_0^{pk_e}\frac{1}{p}\,\mathrm{Im}\left(\frac{1}{\epsilon(p,\omega)}\right) \\ &\quad \times \Theta\left(\frac{k_e^2 - k_F^2}{2} - \omega\right)d\omega dp, \end{aligned}$$

$$\begin{aligned} \beta_2(k_e) = P_{te}^{Pls} &= -\frac{2Z_P^2}{\pi k_e}\int_0^{+\infty}\int_0^{pk_e}\frac{1}{p}\,\mathrm{Im}\left(\frac{1}{\epsilon(p,\omega)}\right) \\ &\quad \times [1 - \Theta(\omega^+(p) - \omega(p))\Theta(\omega(p) - \omega^-(p))] \\ &\quad \times \Theta\left(\frac{k_e^2 - k_F^2}{2} - \omega\right)d\omega dp, \end{aligned} \tag{6}$$

where Z_P is the projectile (proton) charge, $\epsilon(q,\omega)$ is the bulk dielectric response, k_F is the Fermi momentum, and the product of step functions in β_2 both prevents the electron projectile from going into an already occupied state and keeps the calculation outside the binary region [delimited by $\omega^{\pm}(p)=p^2/2\pm pk_F$].

B. Electrons excited by subthreshold plasmon decay

A plasmon excited in the indirect fashion introduced in the previous scenario cannot be itself experimentally identified. The energy-loss method might display evidence of the intermediary electrons excitation but would show no information regarding what those secondary electrons do, once excited. So, in order to allow for some comparison with experimental results some consideration about plasmon decay mechanisms must be added to the theoretical description.

A plasmon excited in the electron gas will eventually decay transferring its energy and momentum to one or a few valence electrons. For low-momentum plasmons, the main plasmon decay mechanism is the excitation of a nearly free electron that performs an interband transition. These electrons can eventually leave the solid and be experimentally detected. Therefore in this section we will consider a proton that excites a nearly free electron via a three-step process involving the excitation of an intermediary electron and an intermediary plasmon. Schematically,

$$H_i^+ + e_i \rightarrow H_f^+ + e_e,$$

$$e_e + \mathrm{FEG} \rightarrow e_f + \mathrm{FEG}^*,$$

$$\mathrm{FEG}^* + \mathrm{NFe}_i \rightarrow \mathrm{FEG} + \mathrm{NFe}_f, \tag{7}$$

where the first step is the excitation of a free electron by the projectile (proton), the second step is plasmon excitation by the excited electron, and the third step is the excitation of a nearly free electron due to plasmon decay. Note that FEG stands for the free electron gas (FEG), FEG* represents a plasmon excited in the FEG, and NFe is a nearly free electron. Also, note that the first two steps are the same already considered in Sec. II A while step 3 accounts for plasmon decay.

In fact, our formalism treats steps 2 and 3 as a unity, considering that the intermediary electron excites a nearly free electron via the excitation and decay of a plasmon.

We proceed to write down the master equations that read

$$\dot{N}_{e,\bar{k}_e}(t) = \alpha(\bar{k}_e)N_0 - \beta_1(k_e)N_{e,\bar{k}_e} - \gamma(k_e)N_{e,\bar{k}_e},$$

$$\dot{N}'_{e,\bar{k}_e} = \gamma(k_e)N_{e,\bar{k}_e}, \tag{8}$$

where $N_{e,\bar{k}_e}$ stands for the number of intermediary electrons in state $\bar{k}_e$, $N'_{e,\bar{k}_e}$ represents the number of nearly free electrons excited by intermediary electrons in state $\bar{k}_e$ via plasmon excitation and decay, and $\gamma(k_e)$ is the probability per unit time for an intermediary electron in state $\bar{k}_e$ to excite a nearly free electron via an intermediary plasmon. Note that, in this treatment, no equation for the number of excited plasmons is considered. The number of intermediary electrons increase due to excitations by the projectile (α), and decrease due to those electrons exciting other electrons, either directly (β_1) or via an intermediary plasmon (γ). This last process will of course increase the number of excited nearly free electrons.

Solving these equations in a fashion analogous to the one illustrated in the previous section, we obtain the probability per unit time for the three-step process P_t^{NFe}. Its final expression is obtained from Eq. (5) just by making the replacements $\beta_2(k_e) \rightarrow \gamma(k_e)$ and $\beta^{tot}(k_e) \rightarrow \beta_1(k_e) + \gamma(k_e)$, where β_1 has already been defined [see paragraph after Eq. (2)], and γ, being the probability of step2+step3 (that is direct plasmon excitation+plasmon decay into a nearly free electron) was obtained in a previous paper [7] where direct plasmon excitation and decay was studied. These coefficients read

$$\begin{aligned}\beta_1(k_e) = P_{te}^{Bin} = -\frac{2Z_P^2}{\pi k_e}\int_0^{+\infty}\int_0^{pk_e}\frac{1}{p}\,\mathrm{Im}\left(\frac{1}{\epsilon(p,\omega)}\right)\\ \times\Theta(\omega^+(p)-\omega(p))\Theta(\omega(p)-\omega^-(p))\\ \times\Theta\left(\frac{k_e^2-k_F^2}{2}-\omega\right)d\omega dp,\end{aligned}$$

$$\begin{aligned}\gamma(k_e) = P_{te}^{\mathrm{NFe}} = 2\pi\int 2\delta\left(\omega - \bar{k}_e\cdot\bar{p} + \frac{p^2}{2}\right)|T_{if}|^2\\ \times\Theta(k_F - k_{1i})\Theta(k_{1f}-k_F)\Theta\left(\frac{k_e^2-k_F^2}{2}-\omega\right)\\ \times[1-\Theta(\omega^+(p)-\omega(p))\Theta(\omega(p)-\omega^-(p))]\\ \times d\bar{p}d\bar{k}_{1i}d\bar{k}_{1f}.\end{aligned} \tag{9}$$

Finally, the transition matrix T_{if} that appears in the expression for γ (obtained by means of a first order Born expansion of the nearly free electron wave function in terms of the lattice potential [4]) is given by

$$\begin{aligned}|T_{if}|^2 = \frac{|\tilde{V}_{P1}^{eff}(\bar{p})|^2}{(2\pi)^3}\sum_{\bar{Q}}\frac{|\tilde{V}_{\bar{Q}}|^2}{\Omega}\delta(\bar{p}-\bar{k}_{1f}+\bar{k}_{1i}+\bar{Q})\\ \times\left|g\left(\bar{k}_{1i},\bar{Q},\frac{|\tilde{V}_{\bar{Q}}|}{\sqrt{\Omega}}\right)+g\left(\bar{k}_{1f},-\bar{Q},\frac{|\tilde{V}_{\bar{Q}}|}{\sqrt{\Omega}}\right)\right|^2,\end{aligned} \tag{10}$$

with $\tilde{V}_{P1}^{eff}(\bar{p}) = 4\pi/[(2\pi)^{3/2}p^2\epsilon_{ML}(p,\omega,\gamma)]$ the target-projectile potential in momentum space, $\tilde{V}_{\bar{Q}}$ the Fourier transformed of the lattice's weak periodic potential (tabulated [8]), $\bar{Q}$ the lattice contribution to the momentum conservation equation, Ω the unit cell volume, and $\epsilon_{ML}(p,\omega,\gamma)$ Mermin-Lindhard dielectric response (γ is the plasmon linewidth for $p=0$). The function $g(\bar{k},\bar{q},\beta)$ reads

$$g(\bar{k},\bar{q},\beta) \equiv \frac{1}{\frac{k^2}{2} - \frac{(\bar{k}+\bar{q})^2}{2} + i\beta}. \tag{11}$$

III. RESULTS

Regarding the expressions for P_t^{Pls} and P_t^{NFe} [Eq. (5) and comments after Eq. (8)], note that our results show an explicit time dependence. In Fig. 1, results for the plasmon excitation probability given by the first of these equations are displayed for different values of the time parameter. It can be clearly observed that, after a fast initial rise, a steady state is achived for $t \geqslant 1000$; therefore we will set $t=1000$ for all our calculations.

Results for the probability are portrayed in Fig. 2 as a function of the proton velocity v_i. Also in that figure, results from our previous study [4,7,8] of direct plasmon excitation (with no intermediary electron) are portrayed in order to establish a comparison with the indirect mechanism studied in this paper. The probability of direct plasmon excitation (dashed line) exhibits the expected theoretical threshold at $v_{thp} \sim 1.24$ a.u. (for proton-Al collisions). The probability of indirect plasmon excitation, via an intermediary fast electron (filled circles joined by a straight line) gives just a minor contribution at high proton velocities. However, at velocities below the threshold, it is the major mechanism of plasmon excitation providing a satisfactory explanation for the mentioned experimental results by Ritzau *et al.* [2].

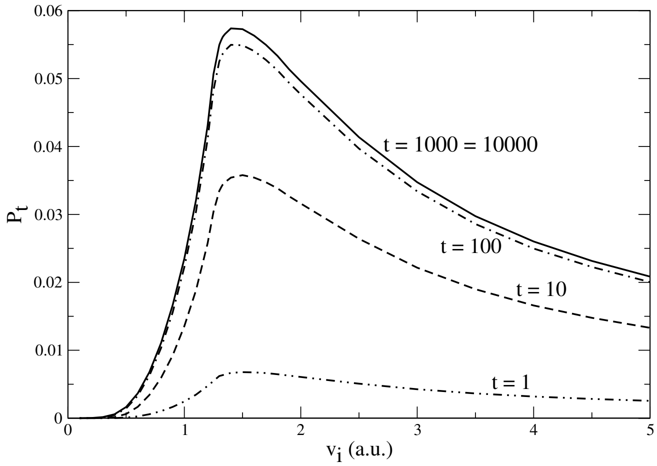

FIG. 1. Probability of indirect plasmon excitation for different values of the time parameter. A steady state is achieved for $t \geqslant 1000$.

Also displayed in Fig. 2, the probability of both direct (dash-dotted line, taken from [7]) and indirect (filled squares joined by a straight line) NFe excitation due to plasmon decay takes our results a step closer to experiments measuring the yield of emitted electrons. We find an important contribution to the NFe excitation yield, at low (subthreshold) projectile velocities, coming from the indirect mechanism.

We now turn to the energy spectrum in Fig. 3, where the probability of nearly free electron excitation due to indirect plasmon excitation and decay is plotted as a function of the nearly free electron final energy for two different projectile velocities $v_i=1.0$ and 1.4 a.u. It is interesting to note that one finds a well-defined peak at an energy $\epsilon_f \approx \omega_P + \epsilon_F$ (zero moment plasmon energy plus Fermi energy). Clearly, this result is not exclusive of subthreshold plasmon excitation and it is also encountered for the direct mechanism [4,7,8]; nevertheless we take the oportunity here to recall that this characteristic of our theoretical results correctly reproduce the energy signature observed in experiments.

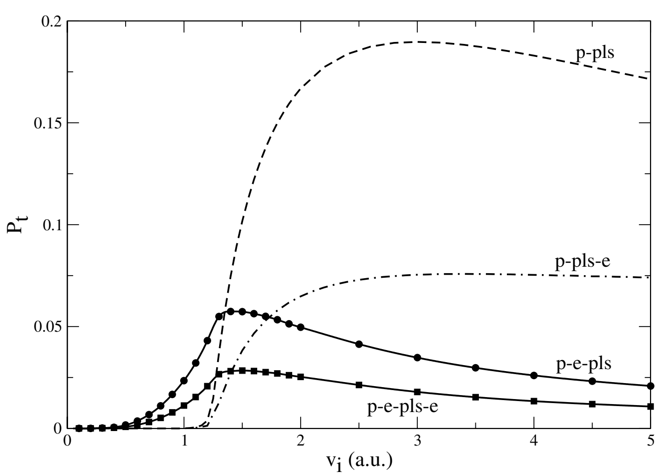

FIG. 2. Probability of plasmon excitation in the FEG both by direct interaction with the proton (*p-pls*, taken from [7]) and with an intermediary electron (*p-e-pls*). Also, probability of NFe excitation via an intermediary plasmon, both with (*p-e-pls-e*) and without (*p-pls-e*, taken from [7]) an intermediary electron.

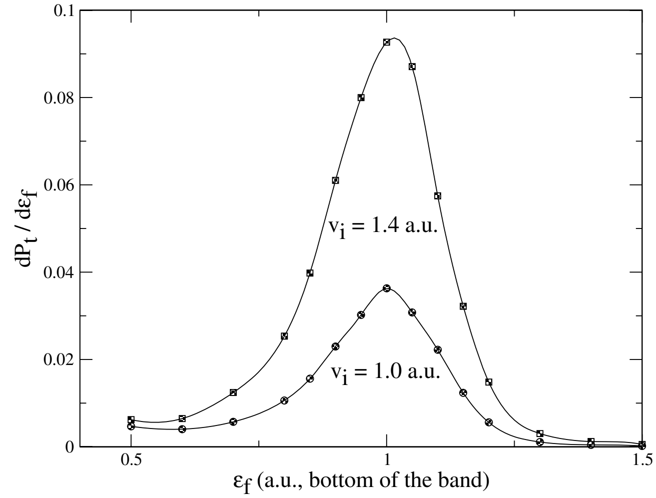

FIG. 3. Energy spectra as a function of the nearly free electron final energy. The process considered is indirect plasmon excitation and later decay via the excitation of a nearly free electron. The peak is located at $\epsilon_f = \omega_P + \epsilon_F$.

Finally, in Fig. 4, a plot is made of dP_t^{NFe}/dk_e as a function of the intermediary electron momentum k_e for a projectile velocity $v_i=1.0$ a.u. This figure gives us information on the momentum distribution of the intermediary electrons who contribute the most to the phenomenon under consideration; we find that it is strongly localized with a peak at $k_e \simeq 1.67$ a.u., above the threshold for electrons $v_{the}=1.55$ a.u. Note that contributions from below the threshold for electrons are due to the finite plasmon linewidth in Mermin-Lindhard dielectric response.

IV. SUMMARY AND CONCLUSIONS

In this paper a theoretical approach to the problem of subthreshold plasmon excitation is presented. An indirect plasmon creation mechanism is studied in which a heavy

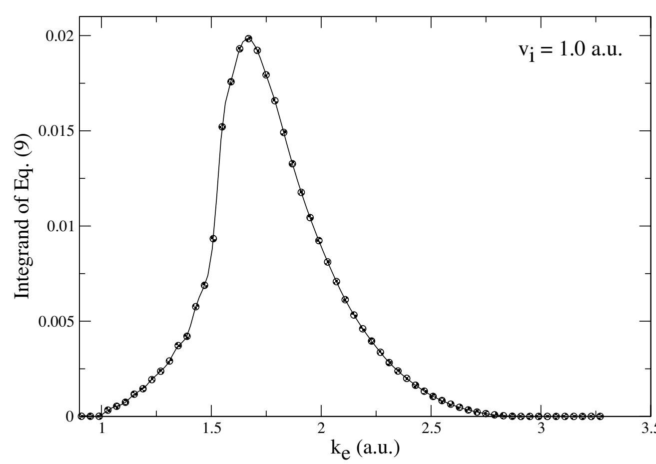

FIG. 4. dP_t^{NFe}/dk_e as a function of the intermediary electron momentum k_e for a projectile velocity $v_i=1.0$ a.u.

projectile (a proton) excites a valence electron that is fast enough to excite a plasmon. The probability of plasmon creation and nearly free electron excitation by plasmon decay are obtained as functions of the projectile velocity.

Also, energy spectra of the excited nearly free electron are displayed for different projectile velocities. The spectra present a clear peak at an energy that coincides with the position of the experimental plasmon shoulder.

The question of the time dependence of our expressions for the probability is briefly addressed and a value for the time parameter is found above which a steady state is achieved.

Finally, some information about the distribution of intermediary electrons capable of exciting plasmons is obtained by plotting dP_t^{NFe}/dk_e as a function of the intermediary electron momentum k_e. We have found that the discussed indirect mechanism of plasmon excitation is non-neglectable for velocities below the threshold. Our results complement the estimates made by Ritzau *et al.* and confirm his conclusions as to the explanation of the measured plasmon excitation by slow ions.

ACKNOWLEDGMENT

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