EXCITATION OF LATTICE MOTION BY INTERACTION WITH LARGE AMPLITUDE ELECTRON PLASMA OSCILLATIONS

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SUMMARY

Fast heavy-ion bombardment on solids in which a "free-electron"-limit for high excitations exists,generates plasma oscillations with very large amplitudes, as found in no other process. This should uniquely enable the observation of their coupling with other degrees of freedom such as phonons. The frequency spectrum of phonons excited by second-order coupling with plasmons and the respective energy loss to the lattice are calculated, using a simple two-fluid-model and the Lagrangian formalism as appropriate for high coherent excitations.

INTRODUCTION

The motivation for this work was the experimental observation of desorption phenomena which are up to now still basically unexplained: fast heavy ion impact on insulator surfaces sets free cluster- (1), atomic (2), and biomolecular (3)(4) secondary ions as well as the respective uncharged particles; the neutral yields typically exceed the ion yields by two orders of magnitude (5). The desorption is observed even in a projectile-velocity-regime where the energy loss is nearly entirely elec-

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tronical, i.e. caused by one-particle and/or collective excitation in the electron system (v≈1cm/ns).

Therefore an explanation for an intensive transfer of energy from the electron system to molecular motion, capable of desorbing clusters or large fragile biomolecules, is of general interest. We concentrated our attention to the excitation of large ordered collective molecular motion with high amplitude. These seem to be required for the desorption of particles with a size above the atomic scale. In view of the very intensive plasmon pulse occuring as one primary process of ion-solid-interaction (6) we tried to investigate the consequences of a coupling between the two collective modes existing in a simple model solid, namely Plasmons and Phonons. The solid was approximated here as assumed to be composed of positively charged ions and a negatively charged fermigas. This ansatz is appropriate to the large coherent excitations encounterd here.

In this model there exist two types of collective excitations: an acustical branch (phonons), charge-neutral and only in weak interaction with a charged projectile, and an optical branch (plasmons) coupling strongly to the projectile (7). Remarkably there is no other process known producing such intensive plasma oscillations as generated here by the interaction with a highly charged fast ion passing a solid; In contrast, in thermal equilibrium such intense excitation amplitudes could not be stimulated without distroying the solid, or the organic molecules, respectively. The approximate independence of desorption of lattice- and electron temperature allows for the the ocurrence even of thermally unstable biomolecules as desorption products and gives a hint as to search for a 'cold' collective surface motion excitation mechanism.

METHOD OF CALCULATION

Assuming that collective excitations play a role in ion-induced desoption phenomena, we can expect observeable effects especially in the large-amplitude-regime, where boson-like excitations can be treated using the classical limit, i.e. using the Lagrangian formalism in our case.

Applying the results in an earlier work of Pines and Schrieffer (7), the interaction between the optical and the acustical mode is established assuming that the electron gas is carried along in lattice motion entirely, thus causing the electrical neutrality of phonons. Then the electron density fluctuates $\propto$-div S (S: lattice displacement field).

The Lagrangian density (LD) is assumed as

$L = T_l + T_e - V_{es} - V_{TF} - V_l$ (F1) with

$T_l = \frac{1}{2} M n v_s^2 (\partial S/\partial t)^2$ (kinetic lattice part),

$T_e = \frac{1}{2} m n \nu v_e^2 j_e^2 / (n\nu(1-\mathrm{div}S))$ (kinetic plasma part),

$V_{es} = \frac{1}{2} m \omega_e^2 \phi_e \, n_e$ (electrostatical part),

$V_{TF} = \alpha (n_e + n\nu(1-\mathrm{div}S))^{5/3}$ (Thomas-Fermi potential part) and

$V_l = \frac{1}{2} M n v_s^2 (\mathrm{div}S)^2$ (elastical lattice part) (F2)

(*with parameters/variables* S: lattice displacement field, j: electron current density, Φ_i, Φ_e: electric potentials due to ion and plasma charge distributions n_e and N_i (without charge dimension), v_s: longitudinal sound velocity, n: density of atoms, M: ionic mass, m: electron mass, α: parameter for TF-energy density, ω_p: plasma frequency, v_F: Fermi-velocity, $v_e^2 = (3/5) v_F^2$).

A series expansion (in powers of $n_e/n\nu$, $j_e/n\nu$ etc.) of this LD yields, in lowest order, quadratic terms; the Lagrangian equations obtained from a LD containing the lowest order terms only are linear wave equations for plasma and lattice motion without an interaction. A further expansion yields higher terms involving interactions between modes "decoupled" in the linear approximation. Here the next higher terms shall be regarded only; these are on the one hand $\frac{1}{2}(\mathrm{div}S) m n \nu v_e^2 (j_e/n\nu)^2$ stemming from the expansion of the kinetical term $\frac{1}{2} m n \nu v_e^2 j_e / (n\nu(1-\mathrm{div}S) - \mathrm{div}j$ and on the other hand $-\frac{1}{2}\mathrm{div}S m n \nu v_e^2 (n_e/n\nu)^2/3$ from the expansion of the Thomas-Fermi-term $\propto n_e^{5/3}$. The LD we have to use is therefore

$$L = \tfrac{1}{2}[Mn((\partial S/\partial t)^2 - v_s^2(\mathrm{div}S)\_^2) +$$
$$+ m n \nu ((j_e/n\nu)^2 - (v_e n_e/n\nu)^2)] +$$
$$+ \tfrac{1}{2} m n \nu v_e^2 (\mathrm{div}S)\Lambda$$

with $\Lambda=-\frac{1}{2}((j_e/v_e n\nu)^2-(n_e/n\nu)^2/3)$. (F3) . The Lagrangian equation for the lattice obtained from this LD is

$v_s^{-2}\partial^2 S/\partial t^2-\Delta S = -\text{grad}\,\Lambda$ (F4) .

To the "free" LD a term representing the action of an external charge moving on the z-axis is added:

$V_p=m\omega_p^2 Z_{eff}^2 n_e/|x-vt|$. (F5) .

(Z_{eff}: effective projectile charge). Direct action of the projectile on the lattice is not regarded here, see (8) for this subject.- In the Lagrangian equations (LE's) for the plasma the phonon interaction term is neglected because the plasma motion is dominated by the projectile action. Then the fouriertransformed potential $\phi_e(\omega,x)$, from which displacement fields, density etc. are obtained by derivation can be calculated analytically by integration with a Greens function (GF). The results thus obtained are used for the calculation of the phonon functions using the respective GF's for the lattice:

$G_l(\omega,y) = \exp(i\omega|y|/v_s)/(4\pi|y|)$ (F6) .

By fouriertransformation $\Lambda(r,\omega=0)=:\Lambda(r)$ is obtained with the z-dependent part (only a phase factor $\exp^2 i\omega z/v^3$) already split off and $\omega=0$ because of the different frequency scales for lattice and plasma motion.

RESULTS

Phonon Excitation

For large distances from the zone of interaction (i.e. where $\Lambda=0$) the fouriertransformed lattice displacement field is $S(\omega,r) = -(\pi/4)\, C(\omega)\, \text{grad}\ \exp\{i\omega z/v\}\, H_o^{(1)}(\omega r/v_s)$ (F7) with

$C(\omega)=\int_o^\infty J_o(\omega r/v_s)\Lambda(r)r\,dr$ (F8) .

The substantial information about the quality of the phonon excitation is involved in the function $\omega C(\omega)$. Fig.2 shows the dependence of $\omega C(\omega)$ of ω for different values of v in units of $[\omega_p k_{TF}^2\ (n\nu\lambda_{TF}^3)]$. The phonon excitation by plasmon-phonon-coupling obviously prefers the low frequency part, i.e. the long waves; this can be explained by the very slow decrease of $\Lambda(\rho)$ for large values of v (see fig.1). One may distinguish two parts which contribute differently to $C(\omega)$: The part stemming from the current term $\propto j_e^2$ is finite for $r=0$ also in the approximation used here, while the density term $\propto n_e^2$ effects a negative contribution prefering the short wave part of the spectrum, due to its logarithmical divergency for $r\to 0$. This contribution is important only for small v; a furhter correction must be applied to this term because the limitation of the plasmon wavenumbers softens the divergent behaviour of $n_e(\omega, r)$. Here the negative values of $\omega C(\omega)$ for small v and larger ω stem from the density term.

Energy Loss to the Lattice by Plasmon-Phonon-Coupling

The energy loss per distance unit dE/dx can be calculated from the radial component of the energy current density *W* resulting from the lattice part of L:

$W = Mnv_s^2(\partial S/\partial t)\,\mathrm{div}\, S$ and

$$(dE/dx)_{pp} = \lim_{R\to\infty} 2\pi R \int_{-\infty}^{\infty} W_r(t,R)\, dt$$

or

$$(dE/dx)_{pp} = (4/\pi)\, Mnv_s^2 \int_0^{\omega_m} (\omega^3/v_s^2)\, |C(\omega)|^2\, d\omega .$$

For this result the properties of the Wronskians of the Besselfunctions were used. For the upper bound of the integral a maximum frequency (here $k_{TF}v_s = \omega_p v_s/v_e$) must be chosen because of the limitation of the frequency spectrum. Furthermore, the divergence of the density term in Λ ($\propto -n_e^2/3$) falsifies the results for small v. Fig.3 shows dE/dx calculated as shown above without correcture (dotted line), calculated neglecting the density term (dashed line) and corrected by replacing r in $\Lambda(\omega=0, r)$ (which is a divergent function for $r\to 0$) by $[r^2+k_{TF}^{-2}]^{-\frac{1}{2}}$ in the interval $0<r<k_{TF}^{-1}$. The correcting term does not cancel the whole contribution (the accuracy of the calculations would be improved by entirely numerical calculating). Nevertheless, a contribution from plasma interactions to lattice motion must also be expected for velocities below v_e (where a resonant plasmon excitation is not possible). In the particle picture this contribution could be regarded as consequence of virtual plasmon exchange.

CONCLUSION

Since the energy loss via plasmon-phonon-coupling is proportional to the ratio electron mass : ionic mass, expressing the different orders of magnitude of the frequency spectra, one cannot expect an action on desorption processes which require large energies. Nontheless the calculations show that the interaction contribution dominates the lattice motion in the high-charge regime: The energy loss calculated from first-order interactions (direct ion-lattice-interaction) is proportional to $(Z_{eff}v_s/v)^2$ which can be compared directly with the function plotted in fig.3 times $Z^4{}_{eff}$.

It is still an open question, how other processes like spike explosion, hot spike explosion or potential modification by electron excitation (9) influence lattice motion. Experimentally the massive production of heavier and fragile fragments indicates that a "cold" energy transport mechanism is required rather than thermal dissipative processes. These latter may however account for the very highest fragments close to the path of the projectile. Explicite calculations of the abundances according to our model for various experimental situations are in preparation. Apart from this our results indicate that phonons excited by plasmon scattering should be observable in experiment. The different time scale of dynamical phonon production by scattering and of quasistatical (i.e. induced by thermodynamical processes in the electron gas) molecule acceleration should enable a registration of the phonons produced by the effect proposed here even in presence of phonons excited by concurring processes.

FIGURES

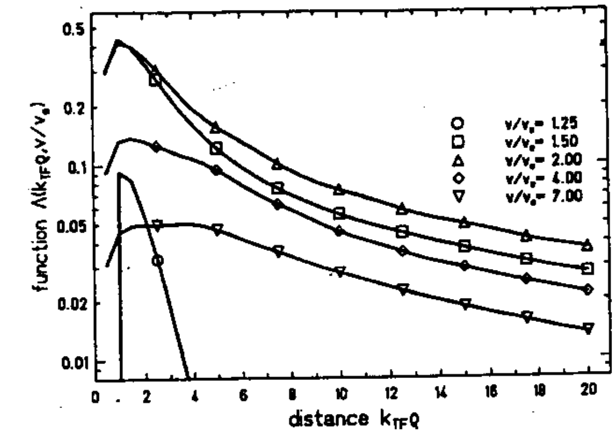

Fig.1: Excitation function Λ in dependence of the distance for different velocities

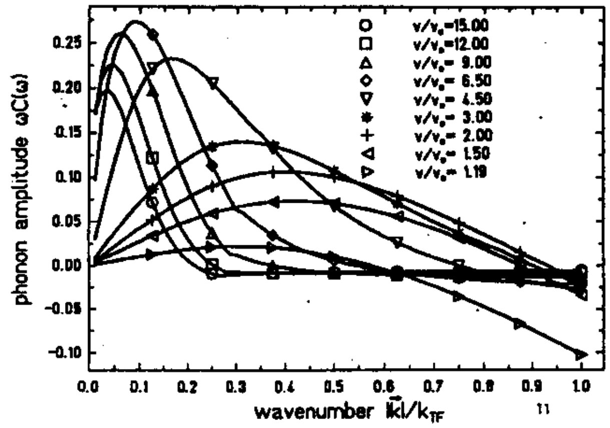

Fig.2: Fouriertransformed phonon amplitude $\omega C(\omega)$ in dependence of the frequency (wavenumber) for different velocities

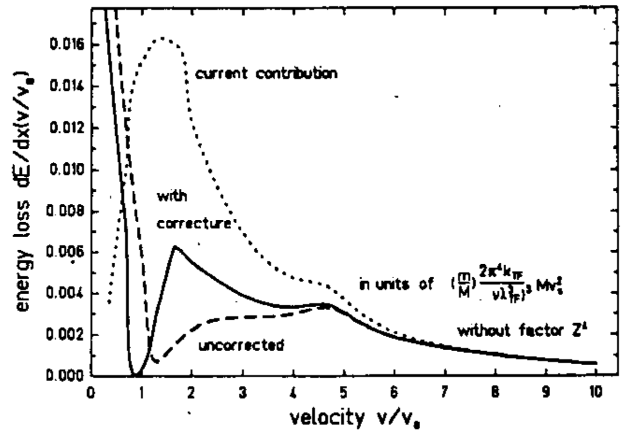

Fig.3: Energy loss to the lattice due to plasmon-phonon-coupling in dependence of the projectile velocity calculated once from the current contribution only (dotted curve), from current and density contribution (dashed curve) and using the short-distance correcture

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