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Collective excitations, optical properties and the stopping power of materials

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ABSTRACT

We compare energy loss by ions and photons *via* electronic excitations in metals, insulators and semiconductors using optical data from synchrotron light-source measurements. The spectra of photon-induced transverse excitations differ markedly from those of longitudinal excitations by ions: photons give up all their energy to a single electronic excitation; charged particles produce a broad spectrum of excitations shifted toward higher energies by collective effects. The widths of these collective-mode resonances in insulators and semiconductors are significantly greater than those in metals reflecting shorter collective-mode lifetimes.

1. Introduction

Energy loss by ions traversing matter occurs principally through electronic excitation. The macroscopic theory of this process [1–9] developed gradually over the middle half of the 20th century from Bohr's microscopic classical theory [10]. The macroscopic theory is based primarily on the matter's complex dielectric response function, $\varepsilon(\mathbf{k},\omega)$, [11] and, while oscillator-model calculations [12–14] have been used to evaluate specific effects, actual measurements of $\varepsilon(\mathbf{k},\omega)$ were not available to test the theory until the development of synchrotron light sources. Here we use optical data[1] to illustrate the processes involved.

Photons and charged-particles lose energy in very different ways[2]. At energies below the onset of Compton scattering, photon energy loss is by photoabsorption. Dipole-allowed transitions dominate and the loss is proportional to $\mathrm{Im}[\varepsilon(\omega)]$. In contrast, swift charged particles lose energy through a series of inelastic electronic excitations each involving a small fraction of the particle's kinetic energy. This is often related to photoabsorption by noting that a passing charge creates a time-varying electric field. The Fourier components of this field may then be considered as a flux of photons ranging from the IR to X-rays. Energy loss occurs by absorption of these virtual photons. However, this picture must be used with care; it does not account for screening by the medium and the incident particle's current. The detailed theory shows that, in contrast to photons, non-relativistic particle energy loss is proportional to $\mathrm{Im}[-1/\varepsilon(\omega)]$ [6,7].

This difference in spectral response is significant for outer-shell electrons. Energy loss by an incident charge to low-lying electronic states is strongly suppressed and all their oscillator strength is shifted to the XUV (10–30 eV). This was implicitly recognized in stopping-power data for cosmic rays [17,18] and was explicitly seen in electron-energy-loss spectroscopy (EELS) of metals [19–21] and insulators [22,23]. The stopping-power effects were explained in terms of dynamic screening by Fermi [4]. In contrast, the EELS experiments were understood within the theory of collective excitations [24–28]. Here we show these are essentially equivalent formulations and illustrate the argument with energy-loss functions calculated from optical data.

Fig. 1 gives these response functions for aluminum, silicon, and vitreous silicon dioxide [29–31]. Particle energy loss to conduction electrons in aluminum displays a sharp collective-mode resonance, or plasmon, but the corresponding resonances in silicon and silica are broad reflecting very short collective-mode lifetimes. These collective effects are significant for outer-shell electrons, but negligible for core electrons that are weakly polarizable (See Figs. 1–3 of [32]).

2. Qualitative theory

Historically, the difference between photon and charged-particle energy loss has been understood in terms of (a) screening, or (b) excitation of longitudinal normal modes of the absorber. The

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[1] Since $\varepsilon(\mathbf{k},\omega)$ is a slowly varying function of $\mathbf{k}$ [15], we neglect the $\mathbf{k}$ dependence and use the frequency-dependent dielectric response derived from optical data corresponding to $\mathbf{k} \approx 0$.

[2] Energy loss to phonons or molecular vibrations is negligible compared with electronic excitations and is neglected. However, it plays a role in energy loss by secondary, tertiary, etc. electrons [16].

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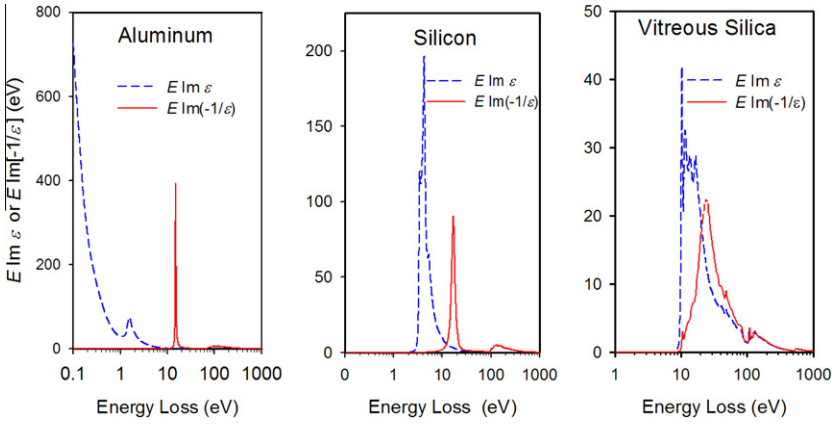

Fig. 1. Energy-loss functions for photons, E Im ε, and charged particles, E Im$[-1/\varepsilon]$, for typical metals, semiconductors and insulators. The energy loss is denoted by $E = \hbar\omega$.

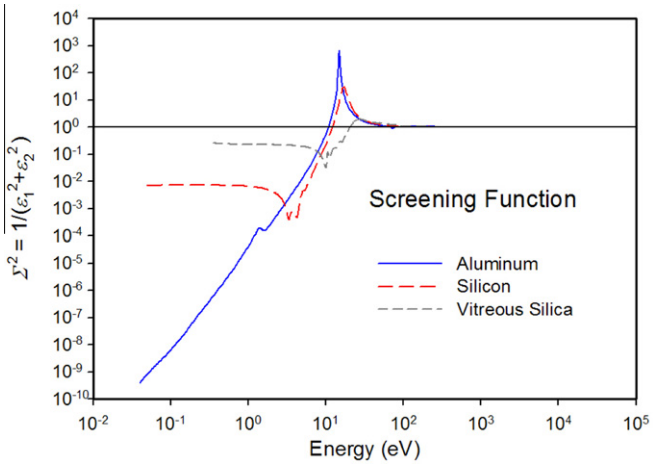

Fig. 2. Screening functions calculated from optical data.

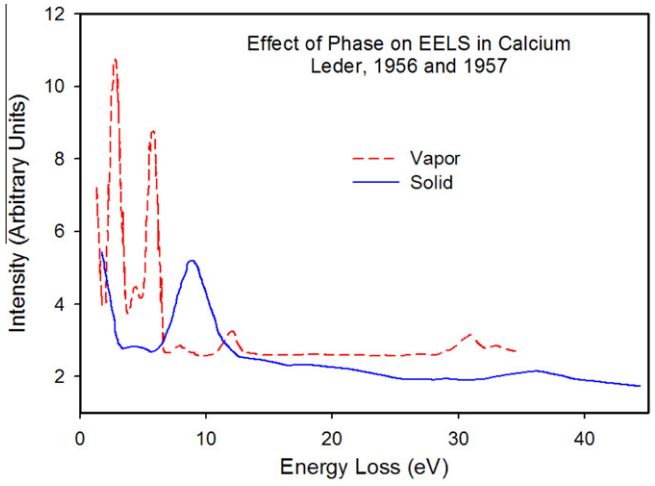

Fig. 3. Electron energy loss in calcium vapor and solid phases after Leder [44].

models are equivalent, but differ in how the Hamiltonian is divided. While detailed treatments of the theory are available [7–9,24,28], qualitative arguments serve to illustrate the physics.

2.1. Screening picture

The screening model is apparent from the microscopic two-body scattering theory [10] by observing that matter between the incident charge and the target particle polarizes and screens the charge. Here, the charged particle and polarization are viewed as coupled and they interact with the absorber as a quasiparticle. Kramers [5] employed this picture to bring von Weizsäcker's theory [33] of energy loss in metals into agreement with experiment. The model is closely related to the dielectric-continuum ideas of Mott, Fröhlich, Pelzer and Hubbard [25–27] for collective excitations. It may be summarized by a plausibility argument based on the different roles played by the electric and displacement fields in Poynting's theorem [34] for the energy density of an electric field in an isotropic dielectric [23,32].

The rate of energy transfer[3] (per unit volume) from the field of the incident particle to the medium *via* dissipation associated with polarization currents induced in the medium is [23,34]

$$\frac{dW}{dt} = -\mathbf{E}\cdot j_{displacement} = -\frac{1}{4\pi}\mathbf{E}\cdot\frac{d\mathbf{D}}{dt}. \tag{1}$$

In the case of a charged particle, the displacement, **D**, is determined by the charge (*via* Div **D**) and the electric field, **E**, follows from $\mathbf{E} = \mathbf{D}/\varepsilon(\omega)$. Neglecting magnetic-field effects, the rate of energy lost by the Fourier component of a particle's electric field with frequency ω is then

$$\left\langle\frac{dW}{dt}\right\rangle \propto -\frac{\omega\varepsilon_2(\omega)}{\varepsilon_1(\omega)^2+\varepsilon_2(\omega)^2}|\mathbf{D}(\omega)|^2 = -\frac{2\pi^2e^2\hbar^2}{m_e}\left(\frac{df_C}{d\omega}\right)|\mathbf{D}(\omega)|^2, \tag{2}$$

where **D** is the independent variable. The quantity $\omega\varepsilon_2/(\varepsilon_1^2+\varepsilon_2^2) = \omega\,\mathrm{Im}[-1/\varepsilon(\omega)]$ measures the energy lost by a charged particle traveling through matter and is often referred to as the energy-loss function for charges. It is proportional to the spectral density of oscillator strength for Coulomb excitations, $df_C/d\omega$, the crucial material property in Bethe's macroscopic theory [2,6–8]. In particular, this spectral density determines the mean excitation energy for Coulomb excitations. (see discussion of Eq. (7) in Section 3.)

In contrast, photons propagate through matter as polaritons [35], i.e., as combined electric and polarization fields. The electric

[3] The sign is chosen so that a decrease in field energy corresponds to a positive energy loss by the particle.