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A study of threshold effects in the energy loss of slow protons in semiconductors and insulators using dielectric and non-linear approaches

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Abstract. The energy loss of slow protons in nonconducting materials, including semiconductors and insulators, is studied using different theoretical methods. First we apply two dielectric models proposed by Brandt and Reinheimer on one side, and by Levine and Louie on the other, and describe in detail the properties of individual and collective contributions according to each model. In addition, we perform an alternative calculation using a non-linear approach based on transport-cross-section methods. These different approaches are compared with experimental results for two semiconductors (Si and Ge) and two insulators (LiF and AlF3), obtaining an approximate description of threshold effects at very low energies. Some interesting similarities and discrepancies are found, which show the current limitations of the theoretical descriptions provided by these methods.

1 Introduction

During the last decades, the interaction of light ions with materials became a tool of interest for several topics of applied physics, from medical treatments to material science and nanotechnology. Moreover, the developments of faster electronics devices based on semiconductor and insulator materials, produced by shallow ion implantation, require a refined knowledge of the properties of the stopping of ions in the range of very low energies, which is the range where band gap effects become more important.

The earlier theoretical models concentrated mostly in the case of swift ions, where inner-shell properties play a significant role, while the specific properties of the valence electron of the target material were not considered in much detail.

First experimental evidence of the so-called threshold effect in the stopping power was obtained by Golser and Semrad in 1991 [1] showing a large deviation of the energy loss of protons in He, in the range of low energies, with respect to common theoretical predictions.

In the case of solid materials, the search for similar effects for several large band-gap materials produced negative results [2–4]; but additional experiments for the same materials showed the existence of threshold effects in the energy loss of protons at very low energies [5–7]. Similar experiments on Si, a semiconductor with a small indirect band gap, showed an almost linear dependence of the stopping power as a function of the ion speed [8]. More recently, energy loss experiments in Ge, showed evidence of a threshold at very low energies [9]. This set of experimental results provide a good framework for the analysis of threshold effects at very low energies in different materials.

We will not consider in this study the cases of several transition metals where similar threshold effects associated to the properties of d electrons were also observed [10–15].

On the theoretical side, several models have been proposed to describe the electronic excitations in semiconductors and insulators using dielectric formulations based on analytical approaches. However a precise comparison between them has never been made, and so the present study fills an important gap in this respect. The range of low energies is a complicated domain and it has been shown that non-linear effects play an important role in the energy loss of even light ions [16–18]. So, one cannot expect linear models to describe accurately the magnitude of the energy loss in this low-energy range. Nevertheless, these models bear significant interest for current studies dealing with semiconductors and insulators, as well as for more elaborate applications to ion stopping in those materials. For instance, several recent studies made use of basic dielectric models (including Lindhard, Mermin [19], and Levine-Louie [20] models) to make a complete analysis of the stopping power of different materials that include valence and inner-shell contributions, and provide a good description of the experimental values for intermediate and high energies. Thus, one may pose the question of to what extent linear dielectric models could describe in

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a reasonable way the existence of threshold effects at low energies, inasmuch as these effects are mainly produced by the dynamical aspects of the interactions, and the ensuing restrictions in the energy transfers to target electrons. This is a problem that has not been yet addressed and so it deserves a detailed and quantitative study.

Based on this view, we aim here to describe in detail the appearance and the characteristics of threshold effects in the interaction of slow protons with semiconductors and insulators from the perspective of dielectric models previously developed for such materials. We based our study on two existing dielectric models [21–23] and we compare the results with those obtained from a non-linear approach based on previous DFT calculations [18] but with additional considerations to include the effect of band gaps in the energy transfer mechanism [24].

The present work is organized as follows: in Section 2 we describe the dielectric approaches used in this study, including a comparison of results on a wide range of energies. In Section 3 we briefly review the non-linear approach based on previous Density Functional calculations and with appropriate considerations to include threshold effects in the energy loss of protons in the range of very low energies. In Section 4 we apply these methods to analyze the threshold effects in semiconductors (Si and Ge) and insulators (LiF and AlF_3), comparing with the available experimental results for these materials. Finally, the conclusions are summarized in Section 5.

2 Dielectric models

We will consider in this work two dielectric models that have been proposed by previous authors: the Brandt-Reinheimer model [21] and the Levine-Louie model [23], showing possible similarities and differences in the results, and concentrating in particular in the effect of the band gaps in semiconductors and insulators. The basic difference between these two dielectric models is the following: the Brandt-Reinheimer model is a full scale calculation of the dielectric response function using first-order perturbation theory, considering transitions matrix elements with the wave functions corresponding to the valence bands of a semiconductor or insulator. On the other hand, the Levine-Louie model is a heuristic approach consistent in a parameterization based on the Lindhard's dielectric function and imposing a displacement of the frequency scale to introduce a gap in the spectrum of excitations. The self-consistency of the formulation is shown by the fulfillment of the sum rules for the energy absorption and for the energy-loss function. To analyze the effects of the energy gap more closely the results of these models will be compared with those of a free electron gas using Lindhard's model [25,26].

2.1 The Brandt-Reinheimer model (BR)

This model is based on previous developments by Callaway [27] and Penn [28]. It has the advantage of showing scaling properties in the dielectric response of the medium, leading also to general scaling properties in the energy loss of the interacting ions.

The dielectric function of the material is expressed in terms of reduced variables in the following way:

$$\varepsilon(k,\omega) = 1 + g(z,E_g)[f_1(z,u,E_g) + i f_2(z,u,E_g)] \quad (1)$$

where k and ω represent the momentum and energy transfers to the medium, and u and z are the corresponding reduced variables defined by the relations: $z = k/2k_F$, $u = \omega/kv_F$. E_g is the energy gap of the material, and v_F and k_F are the Fermi velocity and corresponding wave vector ($k_F = mv_F/\hbar$). Other important quantities to characterize the system are the electron density n, the plasma frequency $\omega_p = (4\pi ne^2/m)^{1/2}$ and the electronic Wigner-Seitz radius $r_s = 1.919/v_F$.

The functions $g(z,E_g)$, $f_1(z,u,E_g)$ and $f_2(z,u,E_g)$ in equation (1) have been calculated in reference [21].

The mean energy loss (stopping power) is calculated in the dielectric formulation by the integral expression:

$$\frac{dE}{dx} = \frac{2}{\pi}\frac{e^2}{v^2}\int_0^\infty \frac{dk}{k}\int_0^{kv} d\omega\,\omega\,\mathrm{Im}\left[\frac{-1}{\varepsilon(k,\omega)}\right] \quad (2)$$

which can be transformed into an integral over the variables u, z using the scaling properties of the model, obtaining

$$\frac{dE}{dx} = \frac{e^2\omega_p^2}{v^2}L(y,E_g) \quad (3)$$

where L is the stopping number, which is a function of the velocity v through the scaling relation (using atomic units for convenience)

$$y = \frac{2v^2}{(\omega_p^2 + E_g^2)^{1/2}}. \quad (4)$$

The stopping number L can be separated in two components, $L = L_{eh} + L_{pl}$, corresponding to the excitation of single individual electrons, or electron-hole pairs (L_{eh}), and collective or plasmon excitations (L_{pl}). The calculation of the eh term is made by integrating equation (2) over the region of the $k-\omega$ plane where the imaginary part of $\varepsilon(k,\omega)$ is different from zero, while the calculation of the plasmon component requires a different procedure; in this case the integral can be transformed into a line integral along the resonance line corresponding to the plasmon dispersion curve defined by $\varepsilon(k,\omega) = 0$. This procedure has been established in reference [26].

In Figure 1 we show some illustrative examples of stopping calculations using the three dielectric models considered here (Lindhard, Brandt-Reinheimer and Levine and Louie). In Figure 1a we show calculations with the Lindhard model, corresponding to a free electron gas, separating the contributions of electron-hole (eh) and plasmon (pl) excitations, and the total stopping given by the sum of both contributions. Here three main aspects of the energy loss phenomenon could be noticed: the proportionality with ion speed in the low energy range, the threshold for plasmon excitation in the region of the stopping power

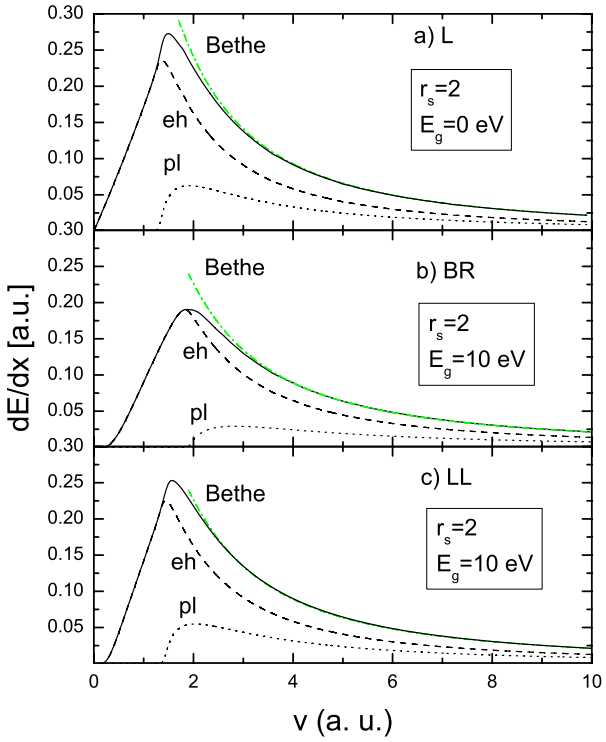

Fig. 1. Stopping power versus the projectile velocity for a proton impinging on a medium represented by a free electron gas with $r_s = 2$ a.u. (a) and with an energy gap $E_g = 10$ eV (b), (c). Dashed-line: individual electron excitations, dotted-line: collective electron excitations, full line: total results, dashed-dotted line: Bethe limit. (a) Lindhard model; (b) Brandt-Reinheimer model; (c) Levine-Louie model.

maximum, and the convergence to the Bethe limit at high energies, i.e.

$$\left.\frac{dE}{dx}\right|_{Bethe} = \frac{e^2\omega_p^2}{v^2}\ln\left(\frac{2mv^2}{\hbar\omega_p}\right). \quad (5)$$

In Figure 1b we show similar calculations using the BR model, for a system characterized by $r_s = 2$ and $E_g = 10$ eV. Here we notice that at low speeds the stopping line deviates from the linear dependence corresponding to a free electron gas. This "threshold effect" is a consequence of the energy gap, which requires a minimum energy transfer $\Delta E = E_g$ to excite electrons across the band gap. For projectile velocities above the threshold the stopping term S_{eh} increases following an approximately linear behavior displaced from the velocity-proportionality, and for still larger velocities, the stopping term S_{eh} reaches a maximum. We notice that the maximum of the stopping power is displaced to larger energies with respect to the Lindhard case, and a more significant displacement occurs in the plasmon term S_{pl}. Finally, for $v \gg v_F$, the total stopping power approaches the asymptotic behavior predicted by Bethe's formula, which in this case reads

$$\frac{dE}{dx} = \frac{e^2\omega_p^2}{v^2}\ln(y) \quad (6)$$

with y given by equation (4).

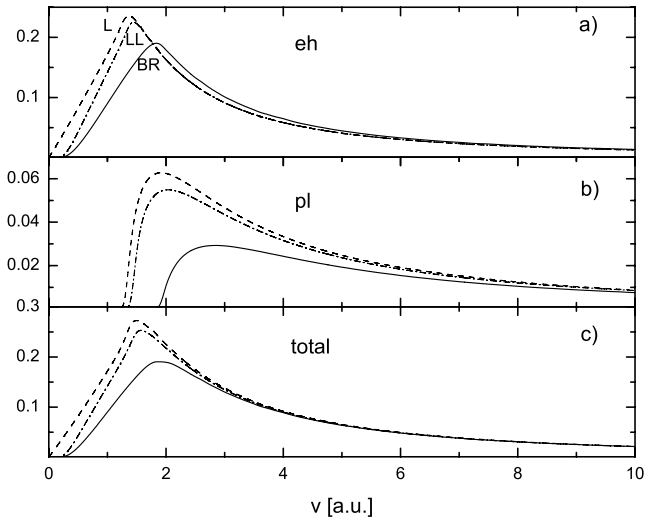

Fig. 2. Separate contributions to the stopping power for the same case of Figure 1. Comparisons between the three dielectric models: dashed-line, Lindhard model, dashed-dotted line, Levine-Louie model, full-line, Brandt-Reinheimer model. (a) Contributions of electron-hole excitations; (b) contributions of plasmon excitations; (c) total stopping values.

2.2 The Levine-Louie model (LL)

This model is directly based on the Lindhard dielectric function for a free electron gas [25], $\varepsilon^L(k,\omega)$, by introducing a displacement in the frequency dependence, of the form

$$\varepsilon_2^{LL}(k,\omega) = \begin{cases} \varepsilon_2^L(k,\omega_-), & \omega \geq E_g/\hbar \\ 0, & \omega < E_g/\hbar \end{cases} \quad (7)$$

where $\varepsilon_2^{LL}(k,\omega)$ and $\varepsilon_2^L(k,\omega)$ are the imaginary parts of the LL and Lindhard models respectively and $\omega_- = (\omega^2 - (E_g/\hbar)^2)^{1/2}$.

The real part of the LL model is obtained from this expression using the Kramers-Krönig relation, which provides an analytical function of k,ω, as described in reference [23].

The stopping power, given by the integral of equation (2), can also be separated into electron-hole (S_{eh}) and plasmon (S_{pl}) components in a similar way as in the BR model.

The calculations of the stopping power using the LL model, for the same material with $r_s = 2$ and $E_g = 10$ eV as before, are shown on Figure 1c. Here the same comments made before regarding the BR calculations apply. Additionally we notice here a more steep increase of the S_{eh} term at low energies, reaching values similar to those in Figure 1a.

In Figure 2 we compare the stopping terms, eh and pl, and the total stopping power obtained from the three dielectric function models for a material with $r_s = 2$, $E_g = 0$ (Lindhard case) and $E_g = 10$ eV (BR and LL cases). Significant differences are obtained, showing more clearly the properties described before. The most significant difference between the BR and LL models is the much larger

shift in the plasmon threshold shown in Figure 2b. This provides and interesting point for future experimental test.

To end this section we notice that all these dielectric functions satisfy the f-sum rule, both with respect to $\varepsilon_2(k,\omega)$ as well as to $\mathrm{Im}[-1/\varepsilon(k,\omega)]$, which is a necessary requirement for appropriate stopping power evaluations.

3 Non-linear model

As it was shown by previous studies [16–18], the range of low energies is a complicated domain where even a proton may be considered a rather strong perturbation when interacting with target electrons in a solid. In such conditions, a non-linear treatment of screening and energy loss is more appropriate. From the point of view of the involved physics, the difference between the non-linear model and the dielectric ones is that in the former case the assumption on the smallness of the perturbation is removed; this produces important differences both in the screening of the external ion potential as well as in the scattering of target electrons by the ion. Thus, the non-linear approach applies also in the cases of very strong screening and large scattering amplitudes, and reduces to the linear treatment when the magnitude of the perturbation is small. The non-linear approach is based on transport cross section (TCS) calculations according to quantum scattering theory. This approach, proposed by Finnemann [29], has been successfully applied to light and heavy ions in metals [18,30–32]. A common feature in the behavior of the stopping power at low energies, according to linear and non-linear predictions, is that the stopping power should be proportional to the ion velocity in the case of simple metals. Deviations from this behavior have been observed in the case of some transition metals, as a consequence of more complicated electronic band-structure properties [24]. A theoretical model has been proposed to take into account the so-called threshold effects in the excitation of nearly-free electrons [24]. This model is based on the transport cross section approach and considers the non-linear aspect of the interactions. With appropriate considerations the TCS approach has been applied in an attempt to describe the threshold effects that arise in the stopping power of protons in wide band gap insulators [6]. To get a more complete perspective of the various approaches, we will include here a description of the threshold effects at low energies using the TCS method, for the present case of semiconductors and insulators.

We restrict the present analysis based on non-linear perturbations to the case of low energies of interest for the applications to be considered next. An extended analysis of non-linear effects in the energy loss of light ions, but without consideration of threshold effects, can be found in reference [33,34].

Using the TCS approach [29], the stopping power in the range of low velocities has the form:

$$\frac{dE}{dx} = nmvv_F\sigma'_{tr}. \tag{8}$$

The restriction in possible energy transfers due to the existence of an energy gap in the target material is imposed in the calculation of the restricted transport cross section σ'_{tr} as follows [24]

$$\sigma'_{tr} = 2\pi \int_{\theta_{\min}}^{\pi} d\theta \sin\theta\,(1-\cos\theta)\,|f(\theta)|^2 \tag{9}$$

where $f(\theta)$ is the scattering amplitude, calculated in terms of the scattering phase shifts δ_l (using for δ_l the values of Ref. [18]), from the well-known expression

$$f(\theta) = \frac{1}{2ik_F}\sum_{l=0}^{\infty}(2l+1)P_l(\cos\theta)\,[\exp(2i\delta_l)-1] \tag{10}$$

and the minimum scattering angle in equation (8) is determined from the condition that the corresponding energy transfer equals the energy of the gap, i.e. [35]

$$\Delta E = mvv_F(1-\cos\theta_{\min}) = E_g. \tag{11}$$

Results obtained from this approach will be discussed in the next section.

4 Applications

We will consider now the energy loss of protons in semiconductors and insulators, with particular attention to the behavior of the stopping powers in the range of very low energies. We will apply the previous dielectric and non-linear models to analyze the effects of the band gaps on the velocity dependence of the energy loss. The aim of these applications is to compare the theoretical predictions with the experimental evidence in various cases where recent measurements are available.

4.1 Semiconductors

We consider here two relevant semiconductor materials, Si and Ge, which were experimentally studied in references [8,9,36]. The indirect band gap energies of these elements are 1.11 eV for Si, and 0.67 eV for Ge.

The results of the calculations using the BR and LL dielectric models, and the non-linear model, are shown in Figure 3, together with the available experimental results of references [8,9,36]. To concentrate on the study of the threshold behavior the theoretical values have been normalized at $v = 0.6$, so that the behavior near the threshold can be more directly tested and compared. In the case of Ge the theoretical models predict threshold velocities in the range of $0.02-0.03$, which must be compared with the experimental value $v_{th} = 0.0274$ according to reference [9]. Considering the dispersion of the experimental points in this energy range, the comparison is fairly good. In the case of Si, the theoretical values of threshold velocities are in the range $0.02-0.04$; the comparison with the experiments in this case is not conclusive due to the differences in the experimental values and because the lowest speeds reached in these experiments are still far from

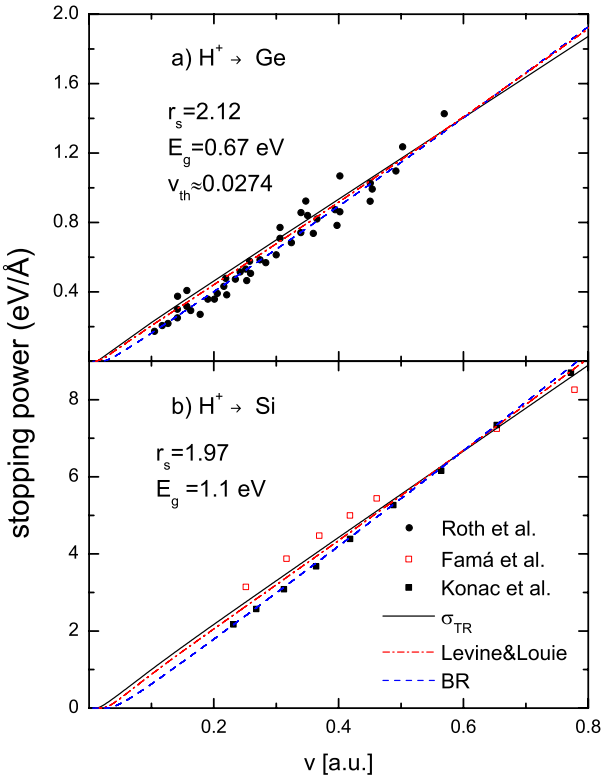

Fig. 3. Stopping power versus the projectile velocity for a proton impinging on two different semiconductors: (a) Ge and (b) Si. Theoretical results from dielectric and non-linear models are compared with experimental results from reference [8] (solid squares), [9] (open squares) and [10] (solid circles).

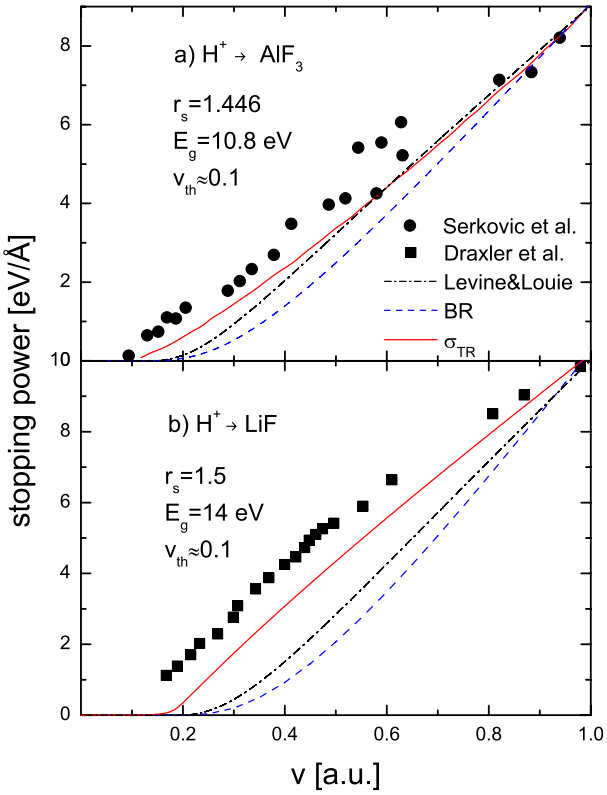

Fig. 4. Stopping power versus the projectile velocity for a proton impinging on two different insulators: (a) AlF_3 and (b) LiF. Theoretical results from dielectric and non-linear models are compared with experimental results from reference [5] (solid squares) and [6] (solid circles).

the predicted threshold values. Hence, we can point out that additional experiments in Si, going down to very low energies, would be important to clarify this question.

4.2 Insulators

Recent experiments have shown the existence of threshold effects in the stopping power of protons, in the range of very low energies, for LiF and AlF_3 [5,6]. The band gap energies for these insulators are 14 eV and 10.8 eV respectively. Therefore one expects to find larger threshold velocities for these materials.

In Figure 4 we compare the theoretical calculations of the stopping powers with the experimental data [5,6]. In this case the theoretical results have been normalized at the highest energy values. The evidence of threshold effects in these insulators is much more clear than in the previous semiconductors. However, the theoretical curves do not reproduce well the trend of the data. The best comparison is obtained in the case of AlF_3 using the non-linear approach.

We also notice that in both insulators the dielectric models predict threshold velocities about twice as large as the experimental values. The reason for this large discrepancy is unknown and intriguing. A possible explanation may lie in an argument provided in reference [2], where a discussion of the distortion of the energy levels in large-band-gap insulators by the presence of a slow proton was given. According to that argumentation, the mechanism of promotion of energy levels would produce an effect that lowers the effective band gap in the vicinity of the proton. This is an interesting problem that may require ab-initio calculations, although it is beyond the scope of the present study.

5 Conclusions

The aim of this study was twofold. One one side we aimed at comparing in detail the results obtained using two dielectric functions of significant interest for stopping power and energy straggling calculations [20]. We find that both theoretical models provide good descriptions of the energy loss process but with some differences in the magnitude of the individual (eh) and collective (pl) contributions. Both dielectric models yield the same velocity threshold, but with a different initial slope; the Brandt-Reinheimer model is the one that shows larger deviations from the results for a free electron gas (Lindhard model), i.e. larger band gap effects. Both models satisfy the f-sum rule and yield the required Bethe limit at high energies, so that both seem equally well qualified for stopping power

calculations. Perhaps the most significant difference between these two models is the larger shift in the plasmon excitation threshold that the BR model predicts. This may be a feature that could allow a more direct and conclusive test of the reliability of each of these models to describe the effects of band gaps in semiconductors and insulators. But for this purpose specific experiments would be required in order to determine the experimental threshold for plasmon excitation by incident protons.

The second objective of this study was to apply these dielectric models, together with a more appropriate non-linear model for slow ions, to attempt to describe the behavior of the stopping power for two semiconductors and two insulator materials, and in particular the threshold effects, in the region of very low energies where experimental results are available. In the case of Si and Ge, the velocity dependence obtained both from linear and non-linear models yield a rather straight-line behavior, in qualitative agreement with the experiments. The theoretical velocity threshold for Ge is in fair agreement with the experiments, while in the case of Si additional experimental determinations for lower energies (i.e. v lower than about 0.2 a.u.) would be required. The results for insulators (LiF and AlF_3) show larger deviations with respect to the experiments and a larger shift in the predicted velocity threshold. The reason for this shift is not currently understood; some previous authors have argued that the presence of an external charge may significantly modify the electronic energy levels, lowering the effective band gap in the vicinity of the moving ion. This argument stands as a plausible physical explanation but requires theoretical investigation in terms of ab initio calculations.

In conclusion, we hope that the present study could stimulate additional experiments and related theoretical work in order to clarify the various aspects indicated before.

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