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Wave-vector-dependent plasmon linewidth in the alkali metals

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The nearly-free-electron (NFE) pseudopotential theory is used to calculate the wave-vector-dependent plasmon linewidth due to interband transitions in lithium, sodium, and potassium. It is shown that interband excitations constitute the dominant decay mechanism which for all alkali metals results in a decreasing linewidth for small $\vec{k}$, contrary to a recent model calculation by Gibbons. Together with estimates of contributions from two particle-hole pair excitations and from phonon- and impurity-assisted intraband and interband transitions, the resulting linewidth is compared with experiment. In Li, the total linewidth decreases for $k \lesssim 0.5k_F$, as borne out by experiment, whereas for Na and K the contribution from pair-pair excitations leads finally to an increasing linewidth, in qualitative agreement with experiment. Remaining quantitative discrepancies suggest that interband transitions to empty d bands beyond the NFE model contribute to the plasmon linewidth in K and possibly also in Na.

I. INTRODUCTION

A study of the homogeneous electron gas within the random-phase approximation (RPA) predicts an infinitely sharp plasmon line, whereas the measured plasmon lines show a wave-vector-dependent linewidth $\Delta E_{1/2}(\vec{k})$ which is characteristic of the system (cf. Raether[1] and references therein). Early theoretical investigations[2,3] concentrated on finding a plasmon decay mechanism due to electron-electron interactions (beyond RPA) within the homogeneous electron-gas model. In order to satisfy momentum and energy conservation such a decay is only possible by two (and higher multiple) electron-hole pair excitations. For the most probable excitation, i.e., the two electron-hole pair excitation, one finds for small $\vec{k}$ the linewidth

$$\Delta E_{1/2}(\vec{k}) = Bk^2 + \cdots . \tag{1.1}$$

First,(1.1) cannot explain the observed finite width for $k \to 0$ and second, the most recent estimates by DuBois and Kivelson[2] and Hasegawa and Watabe[3] show that the coefficient B is too small to account alone for the increase of the linewidth in Al, for example. Since $B > 0$, (1.1) completely fails to explain the peculiar *decrease* of the plasmon linewidth of Li up to $k \sim 0.5k_F$, where k_F is the Fermi wave vector. The decay of the long-wavelength plasmon by phonon- (and impurity-) assisted single particle-hole excitations (where the phonon or the impurity furnishes the necessary momentum), accounted for by a constant relaxation time as in the Drude dielectric function or for finite $\vec{k}$ in the Mermin-Lindhard dielectric function,[4] is neither effective enough to result in a sufficiently large $\Delta E_{1/2}(\vec{0})$ nor does it provide the appropriate variation for finite $\vec{k}$, if the relaxation time is estimated from the dc conductivity.[5]

Although Nozières and Pines[6] suggested early that band-structure effects such as interband transitions should contribute to the plasmon linewidth at $k = 0$, the first calculations of $\Delta E_{1/2}(\vec{0})$ due to interband transitions were only carried out many years later by Paasch[7] for the alkali metals within a nearly-free-electron (NFE) pseudopotential calculation. His results, published as a short note, remained almost unnoticed, as did Hasegawa's[8] rather complicated small-$\vec{k}$ expansion of the interband contribution to $\Delta E_{1/2}(\vec{k})$. In particular, Hasegawa[8] found a decrease of the linewidth for Al and Na, in seeming conflict with experiment. Recently, Sturm[9] confirmed Hasegawa's result for Al but showed that the validity of the k^2 expansion for Al is restricted to a very narrow $\vec{k}$ regime around $k = 0$, where it is practically impossible to observe.

Moreover, by analytically evaluating $\mathrm{Im}\epsilon(\vec{k},\omega)$ due to interband transitions within the NFE pseudopotential approximation for $k < k_c$, the plasmon cutoff wave vector, Sturm[9] was able to explain the observed linewidth for Al within experimental accuracy and also the rather peculiar decrease[10] of $\Delta E_{1/2}(\vec{k})$ for Li. At about the same time, Gibbons[11] performed a model calculation of $\Delta E_{1/2}(\vec{k})$ due to interband transitions in the spirit of an NFE theory for the alkalis. Using a small-$\vec{k}$ expansion, he obtained an increasing $\Delta E_{1/2}(\vec{k})$ for all the alkali metals. His results appear to be in qualitative agreement with experiments in Na and K, but fail in the case of Li.

The purpose of the present work is to apply the NFE pseudopotential theory[9,10] also to Na and K and to demonstrate by a small-$\vec{k}$ expansion why Gibbon's results cannot be correct. As a result we find that $\Delta E_{1/2}(\vec{k})$, due to interband transitions between *nearly-free-electron bands,* decreases for $k \lesssim 0.5k_F$ for Li, Na, and K. This is in principle also true for Rb and Cs; however, the alkali metals, usually regarded as being the closest realization of the model of an (almost) homogeneous electron-gas "metal" or NFE metal which is supported by the observation of almost perfectly spherical Fermi surfaces (apart from Li), show high-frequency optical properties,[12] which in particular for the heavier alkalis appreciably deviate from the expected NFE behavior. As demonstrated for K by a calculation of the optical conductivity from a full band-structure calculation,[13] the additional strong and broad absorption at high frequencies ($\omega \sim 1.5\omega_p$) can be attributed to interband transitions to empty d states. The only exception is Li, where this has not been observed. In Na and K the optical properties around ω_p are well enough below these d-band transitions. For increasing $\vec{k}$, we have to expect contributions from these transitions. Nevertheless, we will assume the validity of the NFE approach and see how far it will take us. In addition, we will show that the contribution from two particle-hole pair excitations in the case of Na and K, when added to the band structure contribution, will result in an increasing linewidth in qualitative agreement with experiment. Contributions from phonon-assisted intraband and interband transitions will be shown to be of minor importance.

II. INTERBAND CONTRIBUTION TO THE WAVE-VECTOR-DEPENDENT PLASMON WIDTH FOR $|\vec{k}| < k_c$

For sufficiently narrow plasmon lines the linewidth $\Delta E_{1/2}(\vec{k})$ can be calculated from

$$\Delta E_{1/2}(\vec{k}) = 2\hbar \left[\mathrm{Im}\epsilon(\vec{k},\omega) \left| \frac{\partial \mathrm{Re}\epsilon(\vec{k},\omega)}{\partial \omega} \right|^{-1} \right]_{\omega_p(\vec{k})} . \tag{2.1}$$

In simple metals local-field effects[14,15] are weak[9] and $\epsilon(\vec{k},\omega)$ can be calculated in the quasihomogeneous approximation.[16] Evaluating $\Delta E_{1/2}(\vec{k})$ to second order in the weak effective crystal potential, $\partial \mathrm{Re}\epsilon(\vec{k},\omega)/\partial\omega$ is replaced by $\partial \mathrm{Re}\epsilon_L(k,\omega)/\partial\omega$. $\omega_p(k)$ is the free-electron plasmon frequency derived from $\epsilon_L(k,\omega) = 0$, where $\epsilon_L(k,\omega)$ is the well-known Lindhard dielectric function.

$\mathrm{Im}\epsilon(\vec{k},\omega)$ for $|\vec{k}| < k_c$ can be calculated analytically[9,10] in the NFE approximation, and one obtains to second order in the crystal potential

$$\mathrm{Im}\epsilon(\vec{k},\omega) = \sum_{\vec{G}\neq\vec{0}} \frac{3\pi}{32} \frac{|U_{\vec{G}}|^2 \omega_p^2}{k^2 k_\perp^2 q} \left[\frac{\psi_1^2}{(1-\psi_1^2)^{1/2}} + \frac{\psi_2^2}{(1-\psi_2^2)^{1/2}} - \frac{(\psi_1+\psi_2)^2}{(1-\psi_1^2)^{1/2}+(1-\psi_2^2)^{1/2}} \right] \times \Theta\left(1 - \left(\frac{\omega - q^2}{2q}\right)^2\right) \tag{2.2}$$

with

$$\psi_{1,2} = 2k_\perp \{1 - [(\omega - q^2)/2q]^2\}^{1/2}/\omega_{1,2} ,$$
$$\omega_{1,2} = \omega \mp k^2 \pm k_{||} q(1 \pm \omega/q^2) , \tag{2.3}$$
$$\vec{q} = \vec{k} + \vec{G},\ k_{||} = \vec{k}\cdot\vec{q}/|\vec{q}|,\ k_\perp^2 = k^2 - k_{||}^2 .$$

Note that in (2.2) all energies are given in units of the Fermi energy $E_F = \hbar^2 k_F^2/2m$ and all wave vectors in units of the Fermi wave vector k_F. From the Θ function in (2.2), it is clear that only those reciprocal-lattice vectors $\vec{G}$ for which $(|\vec{k}+\vec{G}|, \omega_p(k))$ fall into the particle-hole excita-

tion spectrum of the homogeneous electron gas (as illustrated in Fig. 1) will contribute to $\mathrm{Im}\epsilon(\vec{k},\omega)$. It is clear from Fig. 1 that the summation in (2.2) extends only over the first two sets of equivalent reciprocal-lattice vectors for Na. The same can be shown to apply for Li and K. For small $\vec{k}$, i.e., $|\vec{k}| \lesssim 0.5k_F$, only the star of G_{110} contributes and, in the limit $k \to 0$, Eq. (2.2) reduces to the well-known Wilson-Butcher formula, rewritten in the convenient form

$$\mathrm{Im}\epsilon(\vec{0},\omega) = \tfrac{4}{3} n_{G_{110}} \frac{|U_{G_{110}}|^2 G_{110}^4}{\omega^4} \times \mathrm{Im}\epsilon_L(G_{110},\omega) , \tag{2.4}$$

where $n_{\vec{G}_{110}} = 12$ is the number of reciprocal-lattice vectors in the star of G_{110}. The crucial parameter that determines the plasmon linewidth at small $\vec{k}$ is the pseudopotential $U_{G_{110}}$. Estimates of $U_{G_{110}}$ can be derived from experiments that measure Fermi surface properties.[17–19] For $U_{G_{200}}$, one has to rely on estimates based on model potentials.[19,20]

From Fig. 2, where the linewidth for $\vec{k}$ parallel to the three principal symmetry directions [100], [110], and [111] for Li, Na, and K is shown, we observe that for $|\vec{k}| \lesssim 0.5k_F$ in all three cases the linewidth due to interband transitions decreases for increasing $|\vec{k}|$. The reason is simply that the phase space for interband transitions decreases as a

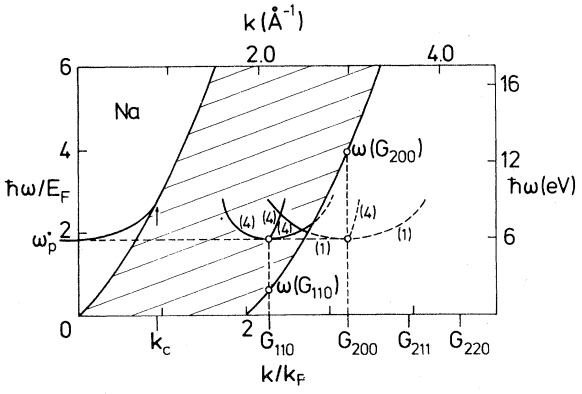

FIG. 1. Homogeneous electron-gas excitation spectrum with density appropriate for Na showing the plasmon dispersion curve and particle-hole continuum (shaded area). Also shown are the loci of the points $(|\vec{k}+\vec{G}|,\omega_p(k))$ with $\vec{k}$ parallel to the [100] direction for $\vec{G} = \vec{G}_{110}$ (12 vectors in the corresponding star) and $\vec{G} = \vec{G}_{200}$ (6 vectors in the star).

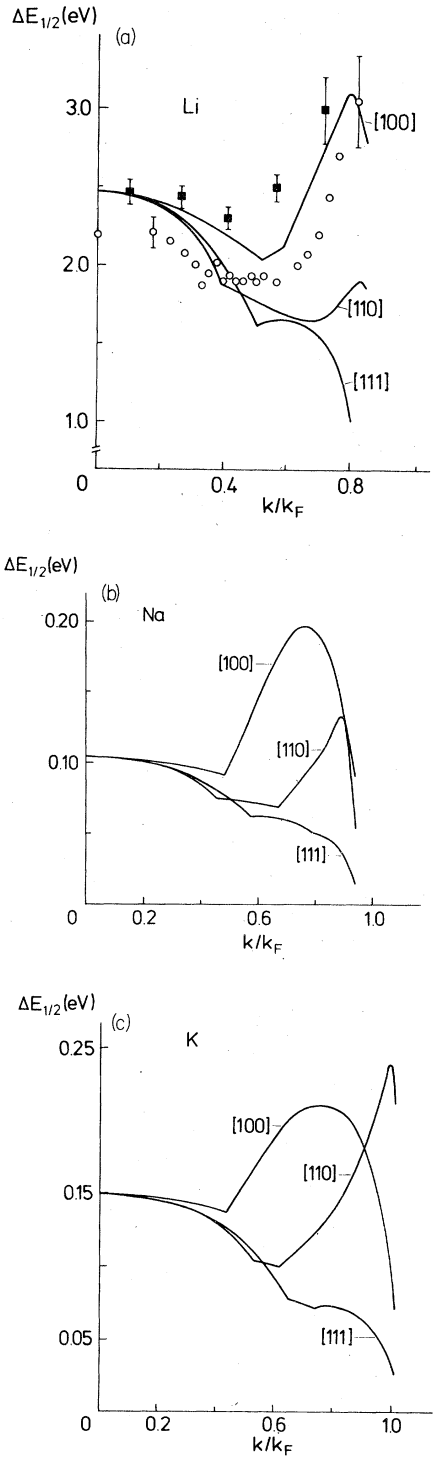

FIG. 2. Plasmon linewidth for (a) Li, (b) Na, and (c) K, calculated from Eqs. (2.1) and (2.2) for $\vec{k}$ parallel to various symmetry directions as indicated. Experimental data for Li were taken from Ref. 21 (circles) and Ref. 22 (full squares).

function of $\vec{k}$ as one can qualitatively understand from Fig. 1. This behavior within the NFE model does also apply to Rb and Cs.

Comparing with experiment, the agreement is particularly very good for Li, which is the real test case because here interband transitions are the most dominant damping mechanism, due to the relatively large $U_{G_{110}}$. For Na and K, interband transitions within the NFE approximation provide roughly half of the observed linewidth, but no decrease for increasing $|\vec{k}|$ is observable. From the strength of the pseudopotential in Na and K one expects a long-wavelength plasmon linewidth of 0.10 and 0.15 eV, respectively, but unfortunately the resolution in most experiments is only of the order of 0.1—0.5 eV for the $\vec{k}=\vec{0}$ linewidth, and it becomes worse for increasing $|\vec{k}|$. Table I displays experimental results for the long-wavelength plasmon linewidth in Li, Na, and K. The results in Table I for Na and K reflect the experimental difficulties well.

Owing to the weak pseudopotential in Na and K, other damping mechanisms such as multiple particle-hole pair excitations, phonon-assisted intraband and interband excitations,and impurity scattering might compete with the interband damping. Before we go into these aspects any further, we want to mention that the present results clearly contradict the results by Gibbons,[11] since he finds a plasmon linewidth due to interband transitions that increases with $|\vec{k}|$ for all the alkali metals. In order to explicitly show where errors must have occurred in the calculation by Gibbons,[11] a small-k expansion is carried out. It also serves to show how well a small-k expansion approximates the result obtained from (2.2). Thirdly, it is also useful for studying the small-k behavior when phonon, impurity, and collective effects are added. These effects will be dealt with in Secs. III and IV.

The small-k expansion of $\Delta E_{1/2}(\vec{k})$ due to interband transitions requires the expansion of $\mathrm{Im}\epsilon(\vec{k},\omega_p(k))$ and of $[\partial \mathrm{Re}\epsilon_L(k,\omega)/\partial\omega]^{-1}_{\omega_p(k)}$. To order k^2 we find

$$\left[\frac{\partial \mathrm{Re}\epsilon_L(k,\omega)}{\partial\omega}\right]^{-1}_{\omega_p(k)} = \frac{1}{2}\left[\omega_p - \frac{\hbar}{m}\alpha k^2\right] + \cdots , \tag{2.5}$$

where $\alpha = 3E_F/5\hbar\omega_p$. Note that (2.5) differs from Eq. (4) of Ref. 11 by the sign in the large parentheses on the right-hand side. We have

$$\mathrm{Im}\epsilon(\vec{k},\omega_p(k)) = \mathrm{Im}\epsilon(0,\omega_p) + \frac{\hbar}{m}\alpha k^2\left[\frac{\partial \mathrm{Im}\epsilon(0,\omega)}{\partial\omega}\right]_{\omega_p} + \frac{1}{2}\sum_j k_i k_j \frac{\partial}{\partial k_i}\frac{\partial}{\partial k_j}\mathrm{Im}\epsilon(\vec{k},\omega_p) . \tag{2.6}$$

The first two terms of (2.6) are readily obtained from (2.4) and the last term of (2.6) is derived from an expansion of $\mathrm{Im}\epsilon(\vec{k},\omega)$:

$$\mathrm{Im}\epsilon(\vec{k},\omega) = \mathrm{Im}\epsilon(0,\omega)\{1 + (k/k_F)^2[\phi_1(G,\omega) + \chi_G(\hat{k})\phi_2(G,\omega)]\} + \cdots , \tag{2.7}$$

TABLE I. Free-electron plasmon frequency ω_p, pseudopotential coefficients U_{110}, long-wavelength limit $\mathrm{Im}\epsilon(\vec{0},\omega_p)$, and plasmon linewidth $\Delta E^{\mathrm{theor}}_{1/2}(\vec{0})$ calculated within the NFE pseudopotential approximation [Eqs. (2.1) and (2.4)]. Also shown are experimentally determined long-wavelength limit plasmon linewidths for the alkali metals.

	ω_p (eV)	U_{110} (eV)	$\mathrm{Im}\epsilon(0,\omega_p)$	$\Delta E^{\mathrm{BS}}_{1/2}(0)$ (eV)	$\Delta E^{\mathrm{expt}}_{1/2}(\vec{0})$ (eV)
Li	8.0	1.5	0.30	2.5	2.2 ± 0.2,[a] 2.5[b] 2.5,[c] 2.6[d]
Na	6.0	0.25	0.017	0.10	0.25,[c] 0.25 ± 0.03,[e] 0.38,[f] 0.45 ± 0.20,[b] 0.4 ± 0.05[a]
K	4.4	0.24	0.034	0.15	0.2,[g] 0.21,[b] 0.25 ± 0.02,[e,i] 0.25 ± 0.05,[l] 0.3 ± 0.05,[a] 0.4,[b] 0.63[f]

[a]Reference 22.
[b]Reference 21.
[c]Reference 23.
[d]Reference 24.
[e]Reference 11.
[f]Reference 25.
[g]Reference 26.
[h]Reference 27.
[i]Reference 28.
[j]Reference 29.

where $G \equiv G_{110}$ and

$$\chi_G(\vec{k}) = \frac{1}{n_{G_{110}}} \sum_{\{\vec{G}_{110}\}} \frac{(\vec{k}\cdot\vec{G})^4}{k^4 G^4} , \tag{2.8}$$

$$\phi_1(G,\omega) = \frac{5E_F}{2E_G} + \frac{5E_F}{\hbar\omega} - \frac{5E_F E_G}{2(\hbar\omega)^2} + \frac{10E_F^2}{(\hbar\omega)^2} + \frac{E_F G}{2E_G}\left[\frac{\partial \,\mathrm{Im}\epsilon_L(G,\omega)}{\partial G}\right]\frac{1}{\mathrm{Im}\epsilon_L(G,\omega)} , \tag{2.9}$$

$$\phi_2(G,\omega) = 3\left[\frac{25E_F}{2E_G} - \frac{5E_F}{\hbar\omega} + \frac{9E_F E_G}{2(\hbar\omega)^2} + \frac{10E_F^2}{(\hbar\omega)^2} + \frac{11E_F G}{2E_G}\left[\frac{\partial \,\mathrm{Im}\epsilon_L(G,\omega)}{\partial G}\right]\frac{1}{\mathrm{Im}\epsilon_L(G,\omega)}\right.$$
$$\left. + \frac{G^2 E_F}{2E_G}\left[\frac{\partial^2 \mathrm{Im}\epsilon_L(G,\omega)}{\partial G^2}\right]\frac{1}{\mathrm{Im}\epsilon_L(G,\omega)}\right] . \tag{2.10}$$

For $\vec{k}$ along [100], [110], and [111], $\chi_G(\hat{k})$ becomes $\frac{1}{6}$, $\frac{5}{24}$, and $\frac{2}{9}$, respectively, and the directional average $\langle \chi_G(\hat{k}) \rangle = \frac{1}{5}$. Using the above equations (2.5)–(2.10) a dimensionless dispersion constant $R(\hat{k})$, defined by

$$\Delta E_{1/2}(\vec{k})/\Delta E_{1/2}(\vec{0}) = 1 + R(\hat{k})(k/k_F)^2 + \cdots , \tag{2.11}$$

can be calculated, which is independent of the effective potential $U_{G_{110}}$. In Fig. 3, the directional average value $\langle R(\hat{k}) \rangle$ as a function of the density parameter r_s is shown to be negative for all alkali metals, which should be compared with the positive values of Gibbon's work.[11]

In Fig. 4, we compare the ratio $\Delta E_{1/2}(\vec{k})/\Delta E_{1/2}(\vec{0})$, due to interband transitions for Li, obtained via the NFE approximation from Eqs. (2.1) and (2.2) with the small-k (or -k^2) expansion from Eq. (2.11). Similar looking figures can be obtained for Na and K. Whereas along the [100] direction the small-k^2 expansion follows closely the "exact" result of the NFE approximation, the difference between the two results increases rapidly with increasing $|\vec{k}|$ and, in particular, the "exact" result along the [110] direction decreases faster (apart from a very narrow regime around $k = 0$) than along the [111] direction in contrast to the small-k^2 expansion. This means that, in principle, the small-k^2 expansion is restricted to a very small $\vec{k}$ regime, $|\vec{k}| < 0.15k_F$ (or $k^2 < 0.02k_F^2$) for Li, Na, and K. But in any case, the important qualitative conclusion from both results is that for all three alkali metals the linewidth due to interband transitions decreases between $k = 0$ and $k \approx 0.5k_F$, contrary to the results by Gibbons.[11] For Li, this behavior is clearly borne out by experiments. For Na and K, however, as will be shown in the next section, additional lifetime effects resulting from the decay of plasmons via two particle-hole excitations will finally result in a total increase of $\Delta E_{1/2}(\vec{k})$ with $|\vec{k}|$.

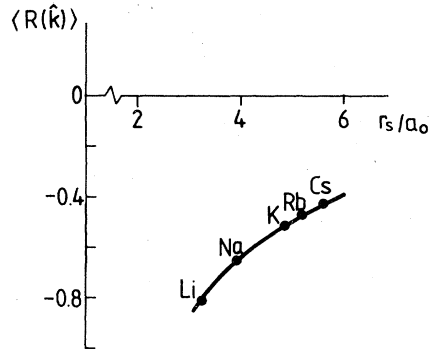

FIG. 3. Dimensionless averaged dispersion constant of the reduced linewidth as a function of r_s. Note that $\langle R(\hat{k}) \rangle$ is by definition [see Eq. (2.11)] independent of $U_{G_{110}}$.

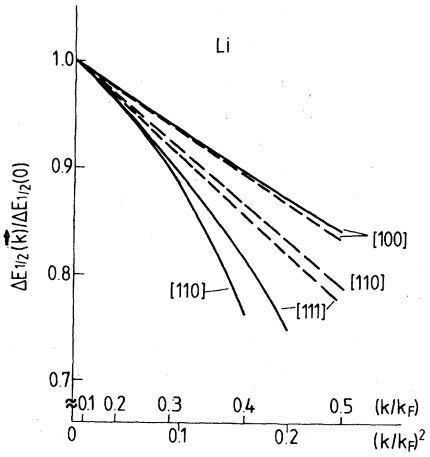

FIG. 4. Reduced plasmon linewidth $\Delta E_{1/2}(\vec{k})/\Delta E_{1/2}(\vec{0})$ for Li. Full curves were obtained from Eqs. (2.1) and (2.2) while the dashed lines represent the small-k^2 result from Eq. (2.11).

III. DECAY OF PLASMONS VIA TWO PARTICLE-HOLE PAIRS

A long time ago, Nozières and Pines[6] discussed the possibility of the decay of plasmons via two particle-hole pairs for finite $\vec{k}$ within the homogeneous electron-gas model beyond RPA. Over the years, various estimates of these contributions have been given by a small-k expansion of $\mathrm{Im}\epsilon^{\mathrm{pair-pair}}(k,\omega)$ [with $\mathrm{Im}\epsilon^{\mathrm{pair-pair}}(0,\omega)=0$]. We adopt here the most recent approximation by Hasegawa and Watabe[3] and DuBois and Kivelson,[2] in which the particle-hole pairs are screened dynamically in the spirit of the RPA, i.e.,

$$\Delta E_{1/2}^{\mathrm{pair-pair}}(k)=\hbar\omega_p\,\mathrm{Im}\epsilon^{\mathrm{pair-pair}}(k)\,, \tag{3.1}$$

where $\mathrm{Im}\epsilon^{\mathrm{pair-pair}}(k)$ is given by

$$\mathrm{Im}\epsilon^{\mathrm{pair-pair}}(k)=b(k/k_F)^2+\cdots \tag{3.2}$$

with

$$b=\frac{1}{40\pi\omega_p^2}\int_0^\infty q^2dq\int_0^{\omega_p}dx\frac{\mathrm{Im}\epsilon_L(q,x)}{|\epsilon_L(q,x)|^2}\left[16(\omega_p-x)^2\,\mathrm{Im}\epsilon_T(q,\omega_p-x)+23\omega_p^2\frac{\mathrm{Im}\epsilon_L(q,\omega_p-x)}{|\epsilon_L(q,\omega_p-x)|^2}\right], \tag{3.3}$$

where wave vectors are given in units of the Fermi wave vector k_F and energies in units of the Fermi energy E_F. The functions $\epsilon_L(q,x)$ and $\epsilon_T(q,x)$ correspond to the longitudinal and transverse Lindhard dielectric functions.[30] Hasegawa and Watabe[3] evaluate the coefficient b for Al and Na, while DuBois and Kivelson[2] performed the calculation for Al. The discrepancies in their numerical estimates of the coefficient b for Al can be traced to a mistake in the expression for $\mathrm{Im}\epsilon_T$ used by DuBois and Kivelson.[2] We calculated b as a function of r_s/a_0, where $r_s=(3/4\pi n_0)^{1/3}$ and n_0 is the valence-electron density, and displayed the result in Fig. 5. It shows, as expected,that the effect is small for high densities and essentially saturates for $r_s/a_0\approx 6$. Adding collective effects (pair-pair excitations), Eqs. (3.1)–(3.3), and band-structure effects (interband), Eq. (2.11), one may write for the reduced plasmon linewidth

$$\Delta E_{1/2}(k)/\Delta E_{1/2}^{\mathrm{BS}}(0)=1+\gamma(k/k_F)^2 \tag{3.4}$$

with

$$\gamma=\langle R(\hat{k})\rangle+\frac{b\hbar\omega_p}{\Delta E_{1/2}^{\mathrm{BS}}(0)}\,. \tag{3.5}$$

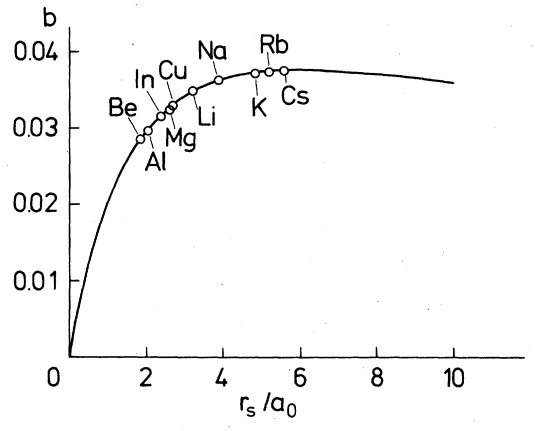

FIG. 5. Coefficient b of $\mathrm{Im}\epsilon^{\mathrm{pair-pair}}(k,\omega_p)=b(k/k_F)^2$, due to the decay of a plasmon into two electron-hole pairs, as a function of r_s/a_0.

Values for γ are found to be $\gamma(\mathrm{Li})=-0.70$, $\gamma(\mathrm{Na})=+1.54$, and $\gamma(\mathrm{K})=+0.58$. In Fig. 6, we compare our theoretical results for $\Delta E_{1/2}(k)/\Delta E_{1/2}^{\mathrm{BS}}(0)$ with the experimental data from Kunz,[21] Kloos,[22] and Gibbons *et al.*[23] for Li, Na, and K. The experimental values used for $\Delta E_{1/2}(0)$ in Fig. 6 are the $k=0$ mean measured values. As results for $\Delta E_{1/2}^{\mathrm{expt}}(0)$ are quite uncertain due to experimental difficulties (see Table I), the reduced plasmon linewidth $\Delta E_{1/2}(k)/\Delta E_{1/2}(0)$ is a useful quantity if one is interested in studying the wave-vector dependence of the linewidth. The theory for Li is in good agreement with the experimental results, the main mechanisms being plasmon decay via interband transitions, e.g., band-structure effects. As is apparent from Fig. 6 for Li, collective effects due to plasmon decay into two particle-hole pairs amount to a small contribution to the wave-vector dependence of the linewidth. For Na, the k dependence of the plasmon width is not very well established experimentally. Our theoretical prediction, however, agrees quite well with the experimental data by Kunz[21] as can be seen from Fig. 6 for

Na. Results in Fig. 6 for Na and K indicate that collective effects lead to an increase with k in the total linewidth in contrast to the predicted decreased obtained if only band-structure effects were taken into account. The theoretical wave-vector dependence of the reduced linewidth for K was found to be 1 order of magnitude less steep than the experimental result by Kloos.[22] Reasons will be discussed in Sec. V.

IV. PLASMON DECAY VIA PHONON AND IMPURITY-ASSISTED INTRABAND AND INTERBAND TRANSITIONS

There is, in general, a small effect at $\omega \sim \omega_p$, and for an estimate we adopt the phenomenological constant relaxation-time approximation, introduced by Mermin,[4] into the Lindhard function in a particle-conserving manner:

$$\epsilon_{\mathrm{ML}}(k,\omega) = 1 + \frac{[1 + i/(\omega\tau)][\epsilon_L(k,\tilde{\omega}) - 1]}{1 + (i/\omega\tau)[\epsilon_L(k,\omega) - 1]/[\epsilon_L(k,0) - 1]} , \tag{4.1}$$

where ML stands for Mermin-Lindhard and $\tilde{\omega} = \omega + i/\tau$, where τ is the constant relaxation time. Using (2.1) we find from (4.1) to order k^2 and to first order in $1/\tau$

$$\Delta E_{1/2}^{\mathrm{ph}}(k) = \frac{\hbar}{\tau} + \frac{16}{15}\frac{\hbar}{\tau}\left[\frac{E_F}{\hbar\omega_p}\right]^2\left[\frac{k}{k_F}\right]^2 . \tag{4.2}$$

We restrict ourselves to Na and K, since in Li interband transitions constitute by far the dominant damping mechanism. By writing

$$\Delta E_{1/2}(k) = \Delta E_{1/2}(0) + B(k/k_F)^2 \tag{4.3}$$

we compare in Table II the contributions to the plasmon linewidth due to electron-lattice [interband transitions, Eqs. (2.11)], electron-electron [two particle-hole pair excitations, Eq. (3.1)], and electron-phonon [phonon-assisted transitions, Eq. (4.2)] interactions. The relaxation time in (4.2) was taken from Ashcroft and Mermin[5] for $T = 273$ K. From Table II, we observe that phonons not only give a fairly small contribution to $\Delta E_{1/2}(0)$, but it remains nearly constant for small k. Consequently, their contribution does not decisively influence the qualitative behavior of the linewidth.

V. DISCUSSIONS AND CONCLUSIONS

We have shown in Sec. II that within the NFE approximation the plasmon linewidth due to interband transtions decreases for $k \lesssim 0.5k_F$ for all alkali metals. The negative sign of $\langle R(\hat{k})\rangle$ follows from (a) the decrease of phase space for interband transitions associated with the star of G_{110} for the bcc crystalline structure (see Fig. 3), and (b) the decrease of the strength[31] of the plasmon pole for increasing k, i.e., of $|\partial\epsilon_L(k,\omega)/\partial\omega|^{-1}_{\omega=\omega_p(k)}$. $\langle R(\hat{k})\rangle$ is most negative for Li which, together with the fact that Li also has by far the largest $U_{G_{110}}$, makes the narrowing of the FWHM for small k due to interband transitions a strong and dominant effect in Li, as compared with the two other damping mechanisms. The situation is different in Na and K: In these cases $\langle R(\hat{k})\rangle$ is smaller and $U_{G_{110}}$ is almost an order of magnitude smaller than that for Li. As a consequence the decrease of the FWHM due to interband transitions is more than compensated by the increase of the linewidth due to two particle-hole pair excitations, as calculated in Sec. III. Phonon effects, estimated in Sec. IV, are of negligible importance for $\Delta E_{1/2}(\vec{k})$ of Li and give a sizable contribution to $\Delta E_{1/2}(0)$ in Na in particular, but contribute very little to the $\vec{k}$ dependence for small k.

Throughout this paper, we treated the three damping mechanisms considered in Secs. II–IV independently, and in the spirit of Mathiessen's rule we added them up to obtain the total linewidth. There is no reason *a priori* that this is justified. Indeed, various possible interference effects between the three mechanisms have been discussed in the literature (see DuBois and Kivelson,[2] Hasegawa,[8] and Garik and Ashcroft[33]). They appear to be very small and we do not expect them to change our conclusions qualitatively in an important way.

Finally, one should also note that the effective pseudopotential is an intrinsically temperature-dependent quantity through the Debye-Waller factor. For Li, which has a fairly high Debye temperature Θ_D, this is of little importance at room temperature, but in Na and K, which have a low Θ_D,

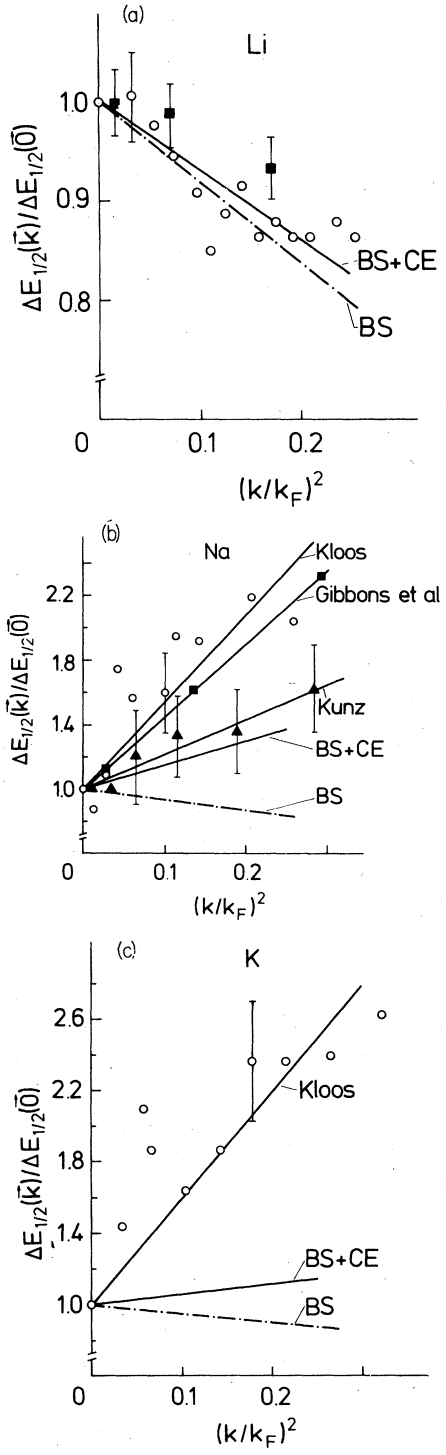

FIG. 6. Reduced plasmon linewidth $\Delta E_{1/2}(\vec{k})/\Delta E_{1/2}(\vec{0})$ for (a) Li, (b) Na, and (c) K. Experimental results are from Ref. 22 (○), Ref. 21 (▲), and Ref. 23 (■). The chained line corresponds to the band structure (BS) result from Eq. (2.11) while the full line labeled BS + CE takes into account band-structure and collective effects according to Eq. (3.5).

TABLE II. Values of $\Delta E_{1/2}(\vec{0})$ and B in Eq. (4.3) due to electron-lattice (interband transitions), electron-electron (two particle-hole pair excitations), and electron-phonon (phonon-assisted transitions) interactions. All values are in eV.

	Na		K	
Interband				
$\Delta E_{1/2}(\vec{0})$	0.10		0.15	
B		−0.065		−0.077
Pair-pair				
$\Delta E_{1/2}(\vec{0})$	0.0		0.0	
B		0.22		0.16
Phonon				
$\Delta E_{1/2}(\vec{0})$	0.021		0.016	
B		0.006		0.004

this will at room temperature reduce the pseudopotential $U_{G_{110}}$ from its low-temperature value as derived from de Haas–Van Alphen effect experiments. Thus the linewidth $\Delta E_{1/2}^{\mathrm{BS}}(k)$ for Na and K must be regarded as an upper limit. Considering the uncertainties (such as energy dependence, for example) in determining the actual $U_{G_{110}}$ at $\omega \sim \omega_p$, we did not include the effect of the Debye-Waller factor. In any case, a decrease of the strength of "direct" interband transitions will occur with increasing temperature with a simultaneous increase of phonon-assisted transitions which results in an increase of $1/\tau_{\mathrm{ph}}$.

Any conclusive comparison with experiment can only be made in the case of Li, and here the agreement is very good both qualitatively and quantitatively (see Fig. 2). For Na and K experimental energy resolution and the linewidth are comparable, allowing hardly any decisive test. From experiment it appears that Na and K have an increasing linewidth at small k in qualitative agreement with our theoretical estimates. Experimentally, for K the linewidth increases much faster, suggesting that the inclusion of transitions to empty d states becomes increasingly important with increasing k. This behavior is beyond the capability of the present NFE approach, in which the pseudopotential is treated as a weak local energy-independent potential and the matrix elements are calculated with plane-wave-like pseudo-wave-functions. Obviously these assumptions become invalid when d states are involved.

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[1]H. Rather, in *Excitations of Plasmons and Interband Transitions by Electrons,* Vol. 88 of *Springer Tracts in Modern Physics,* edited by G. Höhler (Springer, New York, 1980).

[2]D. F. DuBois and M. G. Kivelson, Phys. Rev. 186, 409 (1969).

[3]M. Hasegawa and M. Watabe, J. Phys. Soc. Jpn. 27, 1393 (1969).

[4]N. D. Mermin, Phys. Rev. B 1, 2362 (1970).

[5]N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Holt, Rinehart and Winston, New York, 1976).

[6]P. Nozières and D. Pines, Phys. Rev. 113, 1254 (1959).

[7]G. Paasch, Phys. Status Solidi 38, K 123 (1970).

[8]M. Hasegawa, J. Phys. Soc. Jpn. 31, 649 (1971).

[9]K. Sturm, Z. Phys. B 28, 1 (1977).

[10]K. Sturm, Solid State Commun. 25, 797 (1978).

[11]P. C. Gibbons, Phys. Rev. B 17, 549 (1978).

[12]U. S. Whang, E. T. Arakawa, and T. A. Callcott, Phys. Rev. B 6, 2109 (1972).

[13]W. Y. Ching and J. Callaway, Phys. Rev. Lett. 30, 441 (1973).

[14]S. L. Adler, Phys. Rev. 126, 413 (1962).

[15]N. Wiser, Phys. Rev. 129, 62 (1963).

[16]H. Ehrenreich and M. H. Cohen, Phys. Rev. 115, 786 (1959).

[17]J. J. Donaghy and A. T. Stewart, Phys. Rev. 164, 391 (1967).

[18]M. J. Lee, Proc. R. Soc. London, Ser. A 295, 440 (1966).

[19]N. W. Ashcroft, J. Phys. C 1, 232 (1968).

[20]M. L. Cohen and V. Heine, in *Solid State Physics,* edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1970), Vol. 24.

[21]C. Kunz, Z. Phys. 196, 311 (1966).

[22]T. Kloos, Z. Phys. 265, 225 (1973).

[23]P. C. Gibbons, S. E. Schnatterly, J. J. Ritsko, and J.R. Fields, Phys. Rev. B 13, 2451 (1976).

[24]T. A. Callcott and E. T. Arakawa, J. Opt. Soc. Am. 64, 839 (1974).

[25]H. Mayer and D. Hietel, in *Optical Properties and Electronic Structure of Metals and Alloys,* edited by F. Abelès (North-Holland, Amsterdam, 1966).

[26]N. V. Smith, Phys. Rev. 183, 634 (1969).

[27]P. C. Gibbons and S. E. Schnatterly, Phys. Rev. B 15, 2420 (1971).

[28]R. E. Palmer and S. E. Schnatterly, Phys. Rev. B 4, 2329 (1971).

[29]J. Brambing, Z. Phys. 200, 186 (1967).

[30]J. Lindhard, K. Dans. Vidensk. Selsk. Mat. Fys. Medd. 28, 3 (1954).

[31]We would like to point out an error in previous publications (Refs. 9 and 11), which resulted from an approximate form of $\epsilon_L(k,\omega)$ near $\omega_p(k)$, quoted on p. 150, Eq. (3.180) of Ref. 32,

$$\epsilon_L(k,\omega) \approx 1 - [\omega_p^2(k)]/\omega^2 + i\delta \ ,$$

which leads to the proper dispersion but gives

$$\left| \frac{\partial \epsilon_L(k,\omega)}{\partial \omega} \right|^{-1}_{\omega_p(k)} = \tfrac{1}{2}\omega_p(k) = \tfrac{1}{2}\left[\omega_p + \frac{\hbar}{m}\alpha k^2\right] + \cdots \ ,$$

which obviously increases with k, thus violating an important sum rule.

[32]D. Pines, *Elementary Excitations in Solids* (Benjamin, New York, 1964).

[33]P. Garik and N. W. Ashcroft, Phys. Rev. B 21, 391 (1980).