Lattice-Induced Double Peak in the Dielectric Response of Be Metal

W. Schülke, U. Bonse, H. Nagasawa, S. Mourikis, and A. Kaprolat

Institute of Physics, University of Dortmund, 4600 Dortmund 50, Federal Republic of Germany

(Received 10 March 1987)

Measuring the inelastic scattering of synchrotron x rays by Be metal we found a pronounced double peak in the electronic dynamic structure factor $S(\mathbf{q}, \omega)$ for $\mathbf{q}$ parallel to [110]. We attribute the double peak to the existence of an excitation gap for final states on the (110) Bragg plane. For $q=0.77$ a.u., a zone-boundary collective state could be identified by means of a Kramers-Kronig-deduced zero of $\text{Re}\epsilon(\mathbf{q}, \omega)$. Applications to band analysis are discussed.

PACS numbers: 71.45.Gm, 71.25.Pi, 78.70.Ck

The dielectric response function $-\text{Im}\epsilon^{-1}(\mathbf{q}, \omega)$ and the electronic dynamic structure factor $S(\mathbf{q}, \omega)$ are connected via the well-known fluctuation-dissipation theorem

$$S(\mathbf{q}, \omega) = -(\hbar q^2/4\pi^2 e^2 \rho) \text{Im}\epsilon^{-1}(\mathbf{q}, \omega).$$

Both quantities are of crucial importance, since they describe the response of the electron system of density ρ to an external perturbation with spatial periodicity $2\pi/q$ and frequency ω . For example, the dynamic structure factor and dielectric response function serve as decisive inputs for the correct handling of dynamic screening in band-structure calculations, surpassing the local-density approximation.^{1,2} It seems highly desirable to determine $S(\mathbf{q}, \omega)$ experimentally in a reliable way, since in calculations $S(\mathbf{q}, \omega)$ is known to be extremely sensitive to the mode of treatment of the electron-electron interaction.

Although with semiconductors and transition metals it is conceded that the correct shape of $S(\mathbf{q}, \omega)$ is equally determined by electron-electron interaction and electron-ion interaction, in the case of *s-p* metals the influence of the lattice periodicity is commonly assumed to be of much less importance.³ Accordingly, one-peak, one-shoulder structures or even double-peak structures of $S(\mathbf{q}, \omega)$, as found experimentally with inelastic x-ray scattering at about 5-eV resolution on such different solids as Li, Be, Al, and graphite, have been attributed to correlation effects of the homogeneous electron liquid.^{4,5} Both the conclusions of Ref. 3 and the approximately equal positions of the peak structures on the ω/ω_F scale (ω_F is the Fermi energy) have been the strongest arguments in support of the correlation-effect interpretation. A large number of theoretical papers were prompted by the findings of Ref. 4 (see Schülke *et al.*⁶ for a brief review).

We present here our most recent results of inelastic-scattering experiments on Be single crystals obtained with synchrotron radiation at about 1-eV resolution, and demonstrate that $S(\mathbf{q}, \omega)$ displays a very strong double-peak structure for certain directions of $\mathbf{q}$. In the following we collect arguments in favor of a lattice-induced origin of this double peak. We thus point toward the electron-ion interaction as the source of at least part of

the fine structure in $S(\mathbf{q}, \omega)$ found so far.

We attribute the direction-dependent peak structure of $S(\mathbf{q}, \omega)$ to gaps in the one-particle energy $E(\mathbf{k}')$ of the excited states. The gap arises for $\mathbf{k}'$ on Bragg planes in the extended zone scheme. As already pointed out by Foo and Hopfield⁷ and as thoroughly discussed by Sturm and Oliveira,⁸ the existence of such a gap entails that, within the limits of a two-band model, the excitation energy $\hbar\omega(\mathbf{q}, \mathbf{k}') = E(\mathbf{k}') - E(\mathbf{k})$, with $\mathbf{k}' = \mathbf{k} + \mathbf{q}$, will also exhibit a gap around its free-electron value,

$$\hbar\bar{\omega}(\mathbf{q} = q\hat{\mathbf{g}}, \mathbf{k}' = \mathbf{k}'_{\perp} + \mathbf{g}/2) = \hbar^2 q(g - q)/2m,$$

with $\mathbf{k}'_{\perp} \cdot \mathbf{g} = 0$, provided that $\mathbf{q}$ is parallel to the reciprocal-lattice vector $\mathbf{g}$ that corresponds to the Bragg plane. It follows that $\text{Im}\epsilon(\mathbf{q}, \omega)$ must also have a gap around $\bar{\omega}$, or at least a dip if the two-band model approximation is surpassed. In the response function $-\text{Im}\epsilon^{-1}(\mathbf{q}, \omega)$ such a dip is reproduced, which, however, depending on the reaction of $\text{Re}\epsilon(\mathbf{q}, \omega)$ to the dip in $\text{Im}\epsilon(\mathbf{q}, \omega)$, is certainly shifted somewhat to larger values of ω , thus causing the double-peak structure of $-\text{Im}\epsilon^{-1}(\mathbf{q}, \omega)$ and $S(\mathbf{q}, \omega)$. In exceptional cases, $\text{Re}\epsilon(\mathbf{q}, \omega)$ may have an additional zero within the dip region of $\text{Im}\epsilon(\mathbf{q}, \omega)$, thus leading to a so-called zone-boundary collective state (ZBCS),^{7,8} which enhances the low- ω peak of the double-peak structure.

This presumably lattice-induced fine structure of $S(\mathbf{q}, \omega)$ has already been found by us in Li metal⁶; however, it was much less distinct than in the present case of Be.

We performed the Be experiments at Hamburger Synchrotronstrahlungslabor (DESY) using synchrotron x rays from the storage ring DORIS. The experimental setup, methods of data processing, and normalization were the same as for Li.⁶ The sample single crystal had been grown from 99.99% high-purity Be. The sample thickness was 1.6 mm.

In the following, model calculations based on the quasi-one-pair excitation approximation of Awa, Yasuhara, and Asaki⁹ support our interpretation. In contrast to Ref. 9, we did not use the free-electron polarization function as a starting point, but the equivalent Ehrenreich-Cohen¹⁰ formalism with energy width (life-

time broadening) of one-electron states included as in Ref. 6. We stayed within the limits of a two-band approximation taking into account only one pair $\mathbf{g}_h, \mathbf{g}_{\bar{h}}$ and allowing for the remaining band structure by introducing an effective band mass m^* . We simulated a band-gap variation across the Bragg plane by assuming the potential coefficient V_g belonging to $\mathbf{g}_h$ to vary as $V_g = V_g^0 \exp(-k_{\perp}^2/2\sigma_r^2)$, with $\mathbf{k}_{\perp} \cdot \mathbf{g}_h = 0$. V_g^0 , m^* , and σ_r serve as free parameters of the model and are chosen to fit the experimental data for only *one* value of q . Besides $\mathbf{g}_h$, only m^* determines the energetic position of the dip. Its width and depth are given by V_g^0 and σ_r , respectively.

As in Ref. 6, Coulomb corrections beyond random-phase approximation (RPA) were allowed for both through the momentum-dependent lifetime of quasiparticle states, calculated by means of Quinn's formalism,¹¹ and through the static local-field factor of Utsumi and Ichimaru.¹² For direct comparison with experimental data all calculated dynamic structure factors $S(\mathbf{q}, \omega)$ were convoluted with the experimental 1-eV resolution function.

Figure 1(a) shows the measured $S(\mathbf{q}, \omega)$ for four different $\mathbf{q}$ all parallel to [110] together with corresponding $S(\mathbf{q}, \omega)$'s of polycrystalline Be. All single-crystal curves exhibit a strong double-peak structure with a dip, the position of which is plotted as function of q in Fig. 2. In the following we present arguments for attributing this structure to the existence of an excitation gap for

FIG. 1. (a) Experimental $S(\mathbf{q}, \omega)$: connected dots, Be single crystal, $\mathbf{q} \parallel [110]$; dashed line, polycrystalline Be; solid line, experimental $\text{Re} \epsilon(\mathbf{q}, \omega)$ (Kramers-Kronig transformation) for $\mathbf{q} \parallel [110]$; dotted line, experimental $\text{Re} \epsilon(\mathbf{q}, \omega)$ of polycrystalline Be. (b) Model-calculated $S(\mathbf{q}, \omega)$: solid line, two-band model ($\mathbf{g}_{110}, \mathbf{g}_{\bar{1}\bar{1}0}$), $\mathbf{q} \parallel [110]$, $m^* = 1.11m$, $V_{110}^0 = 0.15$ a.u., $\sigma_r = 0.6$ a.u., lifetime and local-field corrected; dashed line, same as solid line, but empty lattice; dash-dotted line, free-electron RPA.

final states that are located on the (110) Bragg plane. First, there are the results of a corresponding two-band model calculation of $S(\mathbf{q}, \omega)$ based on the pair $\mathbf{g}_{110}, \mathbf{g}_{\bar{1}\bar{1}0}$, $V_{110}^0 = 0.15$ a.u., $m^* = 1.11m$, and $\sigma_r = 0.6$ a.u., as shown in Fig. 1(b) (solid line), which support the excitation-gap interpretation. Also, shown in Fig. 2, there are the calculated dip position (solid line), its position for $m^* = m$ (dashed line), and the position of the excitation gap according to $\hbar \omega = \hbar^2 q(g_{110} - q)/2m^*$ (dash-dotted line).

In order to demonstrate the large extent of the lifetime and local-field corrections to the pure RPA calculations, we have also plotted in Fig. 1(b) the empty-lattice $S(\mathbf{q}, \omega)$ including local field and lifetime (dashed line), and the free-electron RPA (dash-dotted line). It is mainly the momentum-dependent inverse quasiparticle lifetime, with a steep rise at a momentum which makes possible a decay into a plasmon,^{9,11} that is crucial for the overall shape of the spectra.

One might argue that the agreement between an adjustable model calculation and the experiment, as shown in Figs. 1 and 2, is not of much significance. But since V_g^0 , m^* , and σ_r were fitted to the experiment for only *one* q value ($q = 1.09$ a.u.), it is mainly the agreement with respect to the q dependence of both the energetic position (dispersion) and the shape of the spectra (e.g., the depth of the dip) that makes our interpretation of the double peak conclusive. (The deviation of the experimental dip position at $q = 0.77$ a.u. in Fig. 2 from the model calculation might be due to the assumption of a state-independent band mass m^* which is, in effect, an unjustified oversimplification. Setting m^* equal to $1.3m$ for occupied states and $1.15m$ for empty states, respec-

FIG. 2. Dispersion of the dip position for $\mathbf{q} \parallel [110]$: squares, experiment; solid line, model calculated with $m^* = 1.11m$; dashed line, model calculated with $m^* = m$; dash-dotted line, $\hbar \omega = \hbar^2 q(g_{110} - q)/2m^*$, $m^* = 1.11m$; dotted line, $m_{\text{occ}}^* = 1.3m$, $m_{\text{unocc}}^* = 1.15m$.

tively, already leads to a much better agreement, as may be seen from the dotted curve in Fig. 2.)

For $q = 0.77$ a.u., the low- ω peak of the experimental fine structure is found to be a ZBCS, as proved by the occurrence of $\text{Re}\epsilon = 0$ [solid line in Fig. 1(a)], where $\text{Re}\epsilon$ was calculated by means of a Kramers-Kronig transformation of the experimental data. The $q = 0.77$ -a.u. data of polycrystalline Be do not exhibit such an additional zero of $\text{Re}\epsilon$. To the authors' knowledge this is the first unambiguous proof of a ZBCS based on a $\text{Re}\epsilon$ analysis.

The claimed lattice origin of the double-peak structure of the $S(\mathbf{q}, \omega)$ spectra of Be measured with $\mathbf{q}$ parallel to [110] gets its strongest support from the measurements shown in Fig. 3(a) in which we tilted $\mathbf{q}$ away from $\mathbf{g}_{110}$ by angles α towards certain crystallographic directions, as illustrated in the inset of Fig. 3(a). Obviously a tilt of slightly more than 20° is, independent of the direction of tilt, sufficient to cause the dip to fade away. First of all, it is the strong $\mathbf{q}$ -orientation dependence of the double-peak structure that makes its lattice origin rather convincing. But also the good agreement of the experimental data of Fig. 3(a) with the corresponding model calculations as shown in Fig. 3(b) leads to the conclusion that for q parallel to [110], all final states on the $\mathbf{g}_{110}$ Bragg plane cooperate in building the described deep dip. The condition for cooperation is more and more disrupted with increasing α .

In addition to the experimental results shown in Fig. 3(a), we have also observed the fading away of the [110]

dip for other values of $\mathbf{q}$. To make contact with the earlier results of Ref. 4, we show in Fig. 4 for $q = 1.4$ a.u. that a tilting of $\mathbf{q}$ from the [110] orientation to [100] ($\alpha = 30^\circ$) and to [101] ($\alpha = 41^\circ$), respectively, completely destroys the [110] double peak and reveals some new $\mathbf{q}$ -orientation-dependent fine structure. For $q = 1.4$ a.u., intermediate positions were not investigated.

Furthermore, we have also found dips that could be attributed to Bragg-plane-induced excitation gaps in $S(\mathbf{q}, \omega)$ spectra of Be with $\mathbf{q}$ parallel to the following low-index reciprocal-lattice vectors with nonvanishing structure factor: $\mathbf{g}_{100}$, $\mathbf{g}_{101}$, $\mathbf{g}_{102}$, $\mathbf{g}_{103}$, and $\mathbf{g}_{200}$. The dips were not as strongly marked as for $\mathbf{g}_{110}$ and tend to fade out for $q > 1.09$ a.u. A comprehensive analysis of these findings will be given elsewhere.¹³

Summing up, we are led to the following conclusions: (1) For certain directions of $\mathbf{q}$, band-structure effects in $S(\mathbf{q}, \omega)$ of Be are appreciable, especially for values of $q < 1.4$ a.u. (2) There exists a $\mathbf{q}$ -orientation-dependent fine structure of $S(\mathbf{q}, \omega)$ of Be, which can be shown with a high degree of certainty to be related to excitation gaps for final states on Bragg planes. (3) The final states responsible for this fine structure can be located within the k space of the extended zone scheme. Taken together with the knowledge of $\mathbf{q}$, it follows that the corresponding initial states can also be located. Hence, both the energetic position and the dispersion of the discovered fine structure will evolve as a valuable tool, comparable to angular-resolved photoemission, for band-structure analysis. (4) For special cases ZBCS's can be identified by means of Kramers-Kronig transformation of the measured spectra.

FIG. 3. (a) Experimental $S(\mathbf{q}, \omega)$ for $q = 1.09$ a.u.: connected dots, Be single crystals, $\mathbf{q}$ makes an angle α with $\mathbf{g}_{110}$, orientation of $\mathbf{q}$ in the inset; dashed line, polycrystalline Be. (b) Model-calculated $S(\mathbf{q}, \omega)$ for $q = 1.09$: solid line, two-band model ($\mathbf{g}_{110}, \mathbf{g}_{\bar{1}\bar{1}0}$), α same as in (a), parameters same as in Fig. 1(b); dashed line, same as solid line, but empty lattice.

FIG. 4. Experimental $S(\mathbf{q}, \omega)$ for $q = 1.4$ a.u.: Be single crystal, $\mathbf{q} \parallel [110]$, $\mathbf{q} \parallel [100]$, and $\mathbf{q} \parallel [101]$; polycrystalline Be.

One question must be left unanswered in this analysis, namely, whether there remains fine structure of $S(\mathbf{q}, \omega)$ which is exclusively induced by Coulomb correlation, so that it would be present also in the homogeneous electron liquid. The occurrence of a one-peak, one-shoulder structure found in our polycrystalline spectra [see Fig. 1(a)] might be a strong argument in favor of correlation-induced fine structure. Nevertheless, it cannot be ruled out that it is the orientation average of certain lattice-affected fine structure. The $\mathbf{g}_{110}$ -induced fine structure found here contributes less than 5% to the orientation average, so that its effect is not visible in the polycrystalline spectra. The (103) Bragg plane seems to be of much larger influence on the orientation average.¹³

We thank K. Fischer, R. Nubhardt, A. Millhouse, and H. Schenk-Strauss for valuable help with the measurements at DORIS, Hamburger Synchrotronstrahlungslabor (DESY). This work has been funded by the German Federal Minister of Research and Technology under Contract No. 05 334 AX B2.

¹G. Strinati, H. J. Mattauch, and W. Hanke, Phys. Rev. B **25**, 2867 (1982).

²M. S. Hybertsen and S. G. Louie, Phys. Rev. B **30**, 5777 (1984), and Phys. Rev. Lett. **55**, 1418 (1985).

³K. C. Pandey, P. M. Platzman, and P. Eisenberger, Phys. Rev. B **9**, 5046 (1974).

⁴P. M. Platzman and P. Eisenberger, Phys. Rev. Lett. **33**, 152 (1974).

⁵A. Vradis and G. D. Priftis, Phys. Rev. B **32**, 3556 (1985).

⁶W. Schülke, H. Nagasawa, S. Mourikis, and P. Lanzki, Phys. Rev. B **33**, 6744 (1986).

⁷E.-Ni Foo and J. J. Hopfield, Phys. Rev. **173**, 635 (1968).

⁸K. Sturm and L. E. Oliveira, Phys. Rev. B **30**, 4351 (1984).

⁹K. Awa, H. Yasuhara, and T. Asaki, Phys. Rev. B **25**, 3687 (1982).

¹⁰H. Ehrenreich and M. H. Cohen, Phys. Rev. **115**, 786 (1959).

¹¹J. J. Quinn, Phys. Rev. **126**, 1453 (1962).

¹²K. Utsumi and S. Ichimaru, Phys. Rev. B **24**, 3220 (1981).

¹³W. Schülke, H. Nagasawa, S. Mourikis, and A. Kaprolat, to be published.