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Plasmon Dispersion at Large Wave Vectors in Al

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We report high-precision measurements of electron energy-loss scattering intensity, giving the bulk plasmon dispersion $\omega(q)$ for wave vectors extending to $q > q_F$. Some subtleties are identifiable in these data that clarify the effects of multiple scattering. These results are in qualitative agreement with random-phase-approximation concepts, showing smooth dispersion to $2\omega_p$ and $1.4q_F$, where $\omega(q)$ joins the center of the quasiparticle continuum, rather than the dispersionless result obtained previously.

Recent inelastic x-ray[1] and electron[2,3] observations have suggested a possible breakdown in the random-phase approximation (RPA) for the dynamic structure factor $S(q, \omega)$ of a quasifree electron gas. Specifically, these observations indicate that the plasmon energy becomes constant as a function of q for $q > q_c$ (the wave vector cutoff due to quasiparticle interactions). In addition, the x-ray inelastic scattering indicates the occurrence of unexplained peaks[4] near this energy for $q \approx q_F$ to $q \approx 2q_F$, where q_F is the Fermi momentum. Prompted by these observations, some theoretical calculations have also been reported which produce peaks in the spectra.[5] The purpose of this Letter is to inject caution into the acceptance of at least the electron-scattering results for $q \gtrsim q_F$ and to present an alternative explanation for those observations. We first report some features in raw electron-scattering data from aluminum which suggest an explanation in which no serious modification of the RPA concepts appears necessary. We identify subtleties in the treatment of multiple scattering and discuss a procedure which meets these subtleties. For the guidance of theoretical efforts, it appears important to establish the existence of these features in the data, the ensuing subtleties, and an interpretation consistent with present theory. We also hope to prompt a re-examination of the inelastic x-ray scattering in this regime (i.e., $q_c < q < 1.5q_F$).

The data were taken with the Cornell electron microscope/electron spectrometer[6] controlled on-line with a small computer.[7] Computer control permits the use of many scans to compensate for fluctuations in beam intensity, continuous monitoring and correction of the beam direction (which also fluctuates), and variable counting intervals to achieve relatively uniform statistical accuracy over a wide intensity range ($\sim 10^7$). An incident beam of 75-keV electrons with an energy spread of either 0.9 or 1.8 eV and an angular spread of 0.11 Å^{-1} was used. The Al specimens were deposited at room temperature onto NaCl substrates under a vacuum of $\sim 10^{-8}$ mm Hg. They were then floated onto distilled water and picked up on microscope grids. The resulting samples were self-supporting and consisted of ~100 Å crystallites with [111] preferred orientation. Existing electron energy-loss measurements were carried out on similar specimens.

Figure 1 shows electron intensity as a function of momentum transfer q at constant energy loss. Peaks are clearly discernible to 28 eV, fully 6 eV above the previously reported flattening at 22 eV. This observation is not, however, inconsistent with the previous work[2,3] which reported scans as a function of energy loss at constant q. Specifically, a peak relatively nondispersive in energy may not appear in the angular scans, while the peak represented by Fig. 1 may not be readily apparent in the energy scans. We conclude that there is a strong possibility that two peaks exist.

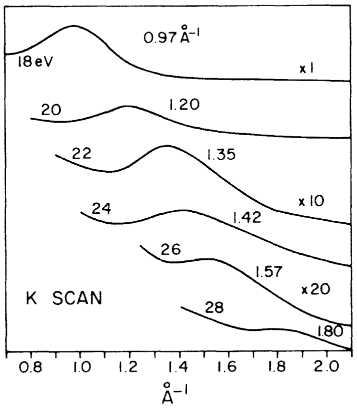

FIG. 1. Scans as a function of scattering angle at fixed energy loss. Peaks are clearly seen to 28 eV, far above the previously reported asymptotic behavior at 22.5 eV.

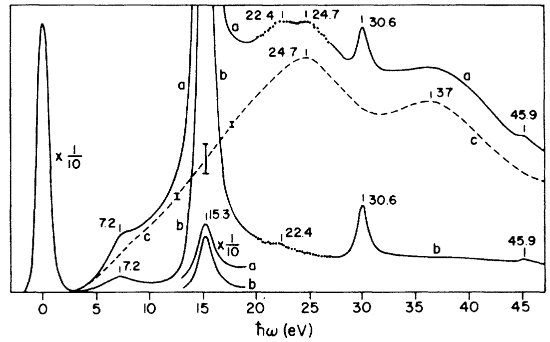

FIG. 2. Detailed correction procedures for $q = 1.8$ Å^{-1}. Curve a shows the measured energy-loss distribution for $q = 1.8$ Å^{-1}. Curve b shows the measured energy-loss distribution for the integrated scattering normalized to curve a at 0 eV. The regions displaying data points were taken with 0.2% statistical accuracy to resolve the peaks between 20 and 30 eV. Curve c shows the difference between a and b and contains only the single and double scattered volume plasmons at 24.7 and 37 eV.

The first is relatively nondispersive at 22 eV, whereas the other is strongly dispersive. It is reasonable to suspect that the strongly dispersive peak may be the plasmon peak, whereas the 22-eV peak may be associated with multiple scattering involving a valence electron ("electronic") scattering event associated with quasielastic (e.g., thermal diffuse) scattering. This is plausible because the quasielastic scattering intensity is typically weakly dependent on scattering angle and occurs at a constant ($\omega = 0$) energy loss. Therefore, we expect relatively dispersionless features to arise from such scattering. A candidate for such a peak is the triple, quasielastic–bulk plasmon–surface plasmon (QPS) scattering event which would occur at an energy of ~22 eV, resulting from the sum of the bulk plasmon energy (~15 eV) and the surface plasmon energy (~7 eV) of an oxidized Al film.

To evaluate this possibility, we have searched for evidence of two peaks in scans of intensity as a function of ω at constant q. Such evidence is not easy to find since the intensity of the dispersive peak varies strongly with scattering angle and specimen thickness. However, for a 500-Å-thick specimen at $q = 1.8$ Å^{-1}, curve a in Fig. 2 shows two such peaks. We note the presence of a quasielastic peak at $\omega = 0$ eV; a quasielastic plus surface plasmon peak at 7.2 eV; and quasielastic plus multiple volume plasmons at 15.3, 30.6, and 45.9 eV. In the region 20 eV $< \omega <$ 28 eV we see not one broad peak as was observed previously, but two peaks, one at 22.4 eV and one at 24.7 eV. At larger q, the intensity of the lower peak remains constant at ~1% of the background while that of the upper peak diminishes rapidly. Also, the position of the lower peak remains constant, while the upper peak disperses rapidly upward. These observations are consistent with the identification of the lower peak as the QPS event, and the upper peak as the single bulk plasmon.

The important question now becomes the quantitative one of separating the two peaks. In estimating the QPS intensity, it must be realized that small but significant contributions at q can arise from the association of quasielastic scattering at points $q - q'$ elsewhere on the Ewald sphere with "electronic" scattering of wave vector q' and energy loss ω. The total intensity for such a process would be $\int Q(q - q')\sigma_D(q', \omega)\, d^2q'$ integrated over the Ewald sphere where $Q(q - q')$ is the quasielastic scattering and $\sigma_D(q', \omega)$ is the differential cross section for all inelastic scattering with electronic scattering the major component. If, as is experimentally the case, one can treat $Q(q')$ as essentially constant, then the energy distribution of this term will be $\sigma_T(\omega) \propto \int \sigma_D(q', \omega)\, d^2q'$ and is shown as curve b in Fig. 2.[8] The following points should be noted. First, the plasmon peak at 15.3 eV agrees closely with the value for the quasielastic plus plasmon peak in the scan at 1.8 Å^{-1} in both intensity and position. Previous attempts[2] at correcting these kinds of effects used $q = 0$ distributions to approximate the forward scattering part of the multiple distribution. We note that, depending on angular resolution, the $q = 0$ plasmon is at 14.9 eV rather than 15.3 eV and that the area underneath this peak is inadequate to account for the quasielastic plus plasmon peak. Next, curve b shows considerable intensity above the 15-eV loss and, further, shows a small but significant peak associated with the plasmon plus surface plasmon loss at 22.4 eV. It is, we believe, the combination of the asymmetric intensity above the plasmon peak together with the small QPS peak that accounts for the nondispersive 22-eV loss. By Fourier transform techniques, we have been able to match the shape of the peak at $\omega = 0$ in the $q = 1.8$ Å^{-1} scan to the shape of the peak at $\omega = 0$ in the integrated scan and show the difference between the scans in curve c. No evidence of the 22.4-eV peak remains, although an increase in the uncertainty in the intensity occurs as indicated by the error bars. All features that we can readily identify as being associated with the quasielastic scattering are cleanly removed. The result leaves the upper peak at 24.7 eV and is further evidence that this is associated with plasmon scattering.

Given data like curve c, i.e., multiple inelastic scattering due only to the valence electrons, it then becomes possible to extract the single scattering as a function of q, ω by use of a technique proposed by Misell and Jones.[9] This technique exploits the relation $\hat{\sigma}_s \approx \ln(1+\hat{\sigma}_m)$ where σ_s is the single-scattering cross section, σ_m, the multiple-scattering cross section, and the caret denotes a Fourier transform in ω, q_x, and q_y. In the result, the bulk plasmon peak can be followed into the region of the particle-hole excitations, showing a decrease in intensity due to the onset of Landau damping as the plasmon crosses the edge of this region at ~1 Å^{-1}. In Fig. 3, we show this behavior giving the dispersion of the plasmon (AB) and single particle peak (CD). Above 2.4 Å^{-1} the single-particle continuum clearly appears to be the dominant scattering and is consistent with the RPA result (CD). The profiles in this region, however, are not accurate enough to verify the existence of the additional peaks reported by Platzman and Eisenberger.[4] We show also the location of the peaks reported by Zacharias (Z); Höchberger, Otto, and Petri (H); and Gibbons, Schnatterly, Ritsko, and Fields (P).[10] It will be noted that Zacharias's data followed the 22.4-eV peak that we identify above as multiple scattering, whereas the data of Höchberger, Otto, and Petrie follow the intensity-weighted average position of these two peaks. The Princeton data follow Zacharias's data up to $q = 1.6$ Å^{-1} but not far enough to show the leveling at 22 eV. Since these data were taken with 300-keV electrons, multiple-

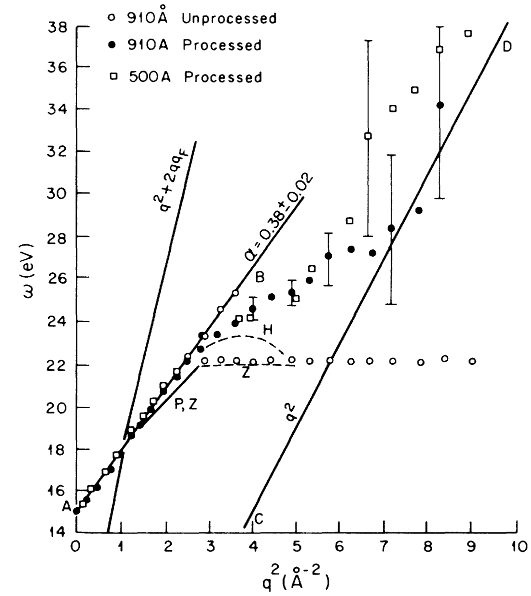

FIG. 3. Results for $\omega(q)$. The dashed lines Z and H denote the results from Refs. 2 and 3, respectively, and the solid line P denotes the result from Ref. 10. Open circles show peak positions in the unprocessed, raw data. Filled circles and open squares show peak positions in the processed data.

scattering effects should be reduced but are not negligible. Assessment of the necessary corrections using our procedures does suggest that these data would then move upwards. Our observations clearly indicate that the bulk plasmon continues to disperse upwards in this region, eventually merging with the single-particle continuum near 28 eV rather than becoming flat at 22.5 eV.

We have been able to reproduce the observed dispersion $\omega(q)$ with a model including the following elements: an RPA Lindhard dielectric constant with $r_s = 2$, a constant plasmon lifetime γ ~0.5 eV introduced following Mermin,[11] a background dielectric constant $\epsilon_B = 1.05$ following Kukkonen,[12] and a q-dependent exchange correction suggested originally by Hubbard[13] but twice as large. A detailed discussion of this will be presented elsewhere but it appears that the unrealistically large values of γ used by Zacharias[2] are not needed for reasonable agreement, if multiple-scattering effects are properly treated.

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Spin Correlations near the Percolation Concentration in Two Dimensions

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Neutron scattering measurements in the dilute antiferromagnets $Rb_2Mn_cMg_{1-c}F_4$ with $c=0.54$ and 0.57 are presented; for this system the percolation limit is $c_p=0.59$. Two-dimensional critical scattering is observed with inverse width ξ and amplitude χT which diverge as a function of $1/T$; the divergences cut off when ξ exceeds the size of the larger clusters. A simple self-avoiding walk model accounts well for the observed ξ vs T behavior.

Recently, considerable attention has been directed towards the percolation transition as an example of "geometrical" critical behavior.[1] Extensive Monte Carlo computer experiments have been performed[2]; in addition, by exploiting an analogy with the Ashkin-Teller-Potts model, some analytical results have been obtained in two dimensions and in $6-\epsilon$ dimensions using renormalization-group methods.[3] As a model percolative system, one may consider a simple square magnet with only nearest-neighbor bonds. As the concentration c of magnetically active atoms is reduced below some critical concentration $c=c_p$, the system breaks up into finite clusters so that there can be no long-range order. Theoretical work to date has concentrated on the critical behavior of such a magnet at $T=0$ around $c=c_p$. More generally, however, the point $c=c_p$, $T=0$ might be considered a *multicritical point* terminating a line of second-order transitions.[4] Such a system then might be expected to exhibit geometrically driven critical behavior at $T\sim 0$ as a function of $c-c_p$ and thermally driven critical behavior at $c=c_p$ as a function of T. Virtually no experimental information is currently available on the T-dependent fluctuation behavior around $c=c_p$ in any real system.

In this Letter we present the results of neutron scattering experiments in the dilute two-dimensional (2D) antiferromagnets $Rb_2Mn_cMg_{1-c}F_4$. The spin correlations in these crystals are found to exhibit a number of interesting and novel features as we shall discuss in detail below. In particular, the data suggest that the propagation of correlations over long distances is determined mainly by the one-dimensional links in the clusters; we show that the principal features of the correlations can be quantitatively accounted for by a no-adjustable-parameter, self-avoiding walk model. In general, we hope that these new results will provide the impetus for the development of theories of percolation in the concentration-temperature plane, and of course, that they will act as a testing ground for such theories.

Our experiments were performed on two high-quality single crystals of $Rb_2Mn_cMg_{1-c}F_4$. These alloys have the Rb_2MnF_4 structure but with the Mn ($S=\frac{5}{2}$) and Mg (nonmagnetic) atoms arranged randomly on the Mn sites of the pure crystal. In the pure crystal the magnetic interactions are