Electron-energy-loss spectroscopy of layered systems

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A feature of electron-energy-loss spectroscopy (EELS) in layered materials is predicted. Contrary to the usual isotropic case, EELS becomes temperature dependent. This result, obtained by using thermodynamic Green's functions, arises from the unusual structure of the layer plasmon bands.

This paper is concerned with electron- and positron-energy-loss spectroscopy (EELS) of layered materials. This well-established experimental technique is very useful in the study of both single-particle and collective excitations such as plasmons in solids (see, for example, Ref. 1). At present, EELS of layered crystals is of particular interest because the high-T_c oxides[2] have a layered structure. In addition to this very topical class of compounds, the approach developed in this paper is applicable to all other types of layered structures such as artificial superlattices, layered materials (graphite), etc.

Recently a semiclassical treatment for a charged particle passing outside a layered material has been given.[3] This paper treats the special case of the energetic particle moving outside the layered material parallel to the layers. The treatment is applicable only at $T=0$ K.

The presence of the layered structure leads to strong modifications of the plasmon spectrum.[4,5] Our goal is to develop a theoretical approach which takes into account the influence of the anisotropic plasmon band on EELS. It will be shown that the unusual structure of the layer plasmon band leads to an experimental loss feature with a characteristic temperature dependence.

Consider the passage of a charged particle through a layered material. The Hamiltonian describing the interaction between the particle and the excitations of the medium has the form

$$H=\sum_{\substack{\mathbf{k},\mathbf{k}'\\ \mathbf{p},\mathbf{q}}} V_{\mathbf{k},\mathbf{p}}^{\mathbf{k}',\mathbf{p}-\mathbf{q}} a_{\mathbf{k}'}^{\dagger}\alpha_{\mathbf{p}-\mathbf{q}}^{\dagger} a_{\mathbf{k}}\alpha_{\mathbf{p}} \ . \tag{1}$$

Here $\mathbf{p}$ is the initial momentum of the particle, $\mathbf{q}$ the momentum transfer, $\mathbf{k}$ and $\mathbf{k}'$ are the initial and final momenta of the material carriers, and

$$V_{\mathbf{k},\mathbf{p}}^{\mathbf{k}',\mathbf{p}-\mathbf{q}}=\int e^{-i(\mathbf{p}-\mathbf{q})\cdot\mathbf{r}_2}\phi_{\mathbf{k}'}^{*}(\mathbf{r}_1)\frac{e^2}{|\mathbf{r}_1-\mathbf{r}_2|}e^{i\mathbf{p}\cdot\mathbf{r}_2}\phi_{\mathbf{k}}(\mathbf{r}_1)d^3r_1 d^3r_2 \tag{2}$$

is the matrix element of the Coulomb interaction between the passing particle and the material carriers. The passing particle, whose wave function is taken as a plane wave, is assumed to be fast ($e^2/hv \ll 1$) and its interaction with the layered material can be treated in the Born approximation (no multiple scattering).

The energy lost by the particle per unit time is equal to

$$\frac{\partial E}{\partial t}\equiv(2\pi)^{-3}\int(\varepsilon_{\mathbf{p}}-\varepsilon_{\mathbf{p}-\mathbf{q}})W_{\mathbf{q}}d^3\mathbf{q} \ , \tag{3}$$

where $W_{\mathbf{q}}$ is the total probability of the transition $\mathbf{p}\rightarrow\mathbf{q}-\mathbf{q}$. In order to evaluate $W_{\mathbf{q}}$, we are adapting the method of Larkin[6] developed for the description of losses in the usual isotropic case to the case of a layered system. A particular advantage of this formulation is that it allows the study of temperature-dependent features.

With the use of the Golden rule and the Hamiltonian [Eq. (1)] we arrive at the following expression for the total probability $W_{\mathbf{q}}$:

$$W_{\mathbf{q}}\equiv 2\pi\sum_{m,n} e^{\beta(\Omega-\mu N_n-E_m)}|R_{mn}(\mathbf{q})|^2\delta(E_m-E_n-\omega) \ , \tag{4}$$

where $\omega=\varepsilon_{\mathbf{p}}-\varepsilon_{\mathbf{p}-\mathbf{q}}$, and

$$R_{mn}(\mathbf{q})=\left\langle m\left|\sum_{\mathbf{k}} V_{\mathbf{k},\mathbf{p}}^{\mathbf{k}+\mathbf{q},\mathbf{p}-\mathbf{q}} a_{\mathbf{k}+\mathbf{q}}^{\dagger}a_{\mathbf{k}}\right|n\right\rangle+R_u \ . \tag{5}$$

Here m,n are eigenstates of the crystal (see, e.g., Refs. 6 and 7); we have separated the term R_u describing Umklapp processes. Equation (4) contains statistical averaging, $\beta=(k_BT)^{-1}$. The matrix element $V_{k',p'}^{k,p}$ is given by Eq. (2). The wave function $\phi_k(r)$ corresponds to the carrier state of the layered material. This function can be written in the form

$$\phi_{\mathbf{k}}(r)=e^{i\boldsymbol{\kappa}\cdot\boldsymbol{\rho}}\chi_{k_z}(z) \ . \tag{6}$$

Here $k=(\boldsymbol{\kappa},k_z)$ is the wave vector of the carrier, and

$$\chi_{k_z}(z)=\sum_{l} u(z-l)e^{ik_z l} \ . \tag{7}$$

The anisotropy of the layered material has been taken into account by writing the function χ_{k_z} in the tight-binding approximation by assuming $u(z-l)$ is localized in the lth layer. With the use of Eqs. (2), (6), and (7) we obtain

$$V_{\mathbf{k},\mathbf{p}}^{\mathbf{k}',\mathbf{p}'}=\frac{e^2}{(\kappa^2+q_z^2)}\int dz\, e^{-iq_z z}\chi_{k_z}^*(z)\chi_{k_z}(z)\ . \tag{8}$$

Substituting (8) into (5) and neglecting the overlap of carrier functions located on different layers (this approximation is not essential as small overlap contributions can be included as shown in Ref. 5), we find

$$V_{\mathbf{k},\mathbf{p}}^{\mathbf{k}',\mathbf{p}'}=V_{\kappa,q_z}\delta_{k_z k_z'+q_z+2\pi g}\ , \tag{9}$$

where

$$V_{\kappa,q_z}=\frac{e^2}{\kappa^2+q_z^2}f(q_z)\ , \tag{10}$$

$$f(q_z)=\int dz|u(z)|^2 e^{-iq_z z}\ . \tag{11}$$

Here $q=|\mathbf{p}-\mathbf{p}'|$, $q_z=p_z-p_z'$, $g=1/c$ describes the contribution from Umklapp processes.

As a result we arrive at the following expression for the total probability W_q [see Eqs. (4), (5), and (9)]:

$$W_{\mathbf{q}}=2\pi|V_{\kappa,q_z}|^2\Phi_{\mathbf{q}}(\varepsilon_{\mathbf{p}}-\varepsilon_{\mathbf{p}-\mathbf{q}})\ , \tag{12}$$

where V_{κ,q_z}^2 is given by Eqs. (10), (11), and

$$\Phi_{\mathbf{q}}(\omega)=\sum_{m,n}e^{\beta(\Omega-\mu N_m-E_m)}\left|\left\langle m\left|\sum_{\mathbf{k}}a_{\mathbf{k}+\mathbf{q}}^{\dagger}a_{\mathbf{k}}\right|n\right\rangle\right|^2 \times\delta(E_n-E_m-\omega)+\phi_{\mathbf{q}}^{u}\ . \tag{13}$$

The function $\Phi_q(\omega)$ has been introduced in Ref. 6. This function can be expressed in terms of the thermodynamic Green function:

$$G(\mathbf{r}_1,\tau_1;\mathbf{r}_2,\tau_2)=\mathrm{Tr}e^{\beta(\Omega-\mu N-H)}T[\hat{n}(\mathbf{r}_1,\tau_1)\hat{n}(\mathbf{r}_2,\tau_2)]\ . \tag{14}$$

Here $\hat{n}$ is the carrier number operator and τ is an imaginary time. According to Ref. 6 the function $\Phi_{\mathbf{q}}(\omega)$ can be written in the form

$$\Phi_{\mathbf{q}}(\omega)=\pi^{-1}\mathrm{Im}\tilde{G}(\mathbf{q},\omega)(1-e^{-\beta\omega})^{-1}\ . \tag{15}$$

The function $\tilde{G}(\mathbf{q},\omega)$ is directly related to the function introduced in Ref. 6. Namely,

$$G(\mathbf{q},2\pi inT)=G_n(\mathbf{q})\ ,$$

where $G_n(\mathbf{q})$ is the Fourier component of the G function.

The thermodynamic Green function $G_n(q)$ can be evaluated with the use of a diagrammatic technique (see Refs. 6 and 7). This evaluation can be carried out in the random-phase approximation (RPA) and as a result, we obtain (cf. Ref. 6)

$$\frac{\partial E}{\partial t}=(4\pi^{-3})^{-1}\int\omega|V_{\kappa,q_z}|^2(1-e^{-\beta\omega})^{-1} \times\mathrm{Im}[\Pi^{-1}(\mathbf{q},\omega)-I_{\kappa,q_z}]d^3q\ . \tag{16}$$

Here $\omega=\varepsilon_{\mathbf{p}}-\varepsilon_{\mathbf{p}-\mathbf{q}}$, $d^3q=q_\parallel dq_\parallel d\phi\, dq_z$, and the matrix element V_{κ,q_z} is defined by Eq. (10). $\Pi(\mathbf{q},\omega)$ is the polarization operator for the layered material and I_{κ,q_z} is the matrix element of the Coulomb interaction between two carriers in the material. In contrast to the isotropic three-dimensional (3D) case it should be noted that $V_{\kappa,q_z}\neq I_{\kappa,q_z}$. The matrix element I_{κ,q_z} can be easily evaluated with the use of Eqs. (6) and (7), but we do not need its explicit form for our present purposes.

Let us evaluate the integral (16). We start with a transformation of variables $\phi,q_z\rightarrow\omega,\Omega_{\mathrm{pl}}(\kappa,q_z)$ (Ω is the layer plasmon frequency). From the conservation of energy, we find

$$\omega=\varepsilon_{\mathbf{p}}-\varepsilon_{\mathbf{p}-\mathbf{q}}=v_\parallel q_\parallel\cos\phi+v_z q_z-q_\parallel^2/2m^*-q_z^2/2m^*\ . \tag{16a}$$

The term $\mathrm{Im}(\Pi^{-1}-I)^{-1}$ is proportional to the imaginary part of the dielectric function of the layered system. The dominant contribution to the integral arises from the poles of the function $(\Pi^{-1}-I)^{-1}$. These poles correspond to the layer plasmon eigenvalues, which have been shown to have the following form in these materials (see Appendix):[4,8]

$$\Omega_{\mathrm{pl}}(\kappa,q_z)=v_F\kappa\left[1+\left[\frac{2F}{r_B\kappa}\right]^2\frac{1}{1+\dfrac{4F}{r_B\kappa}}\right]^{1/2}\ , \tag{17}$$

where

$$F(\kappa,q_z)=\frac{\sinh(\kappa c)}{\cosh(\kappa c)-\cos q_z c}$$

and the effective Bohr radius r_B is given by

$$r_B=\hbar^2\varepsilon_M/m^*e^2\ . \tag{18}$$

As was discussed in our earlier papers[8] the plasmon density of states is sharply peaked at $q_z=0,\pi/c$. c is the interlayer distance. As a result, the derivative $\nabla q_z/\nabla\Omega$

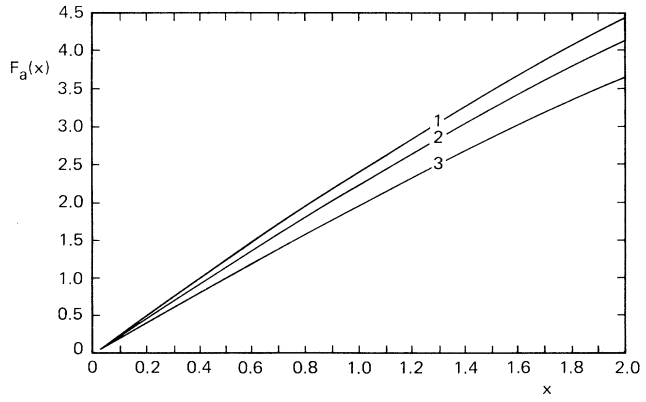

FIG. 1. The function $F_a(x)$: the curves 1,2,3 correspond to values of $a=5.17$ Å ε_M and 4.5 ($s=2.5v_F$), $a=4.34$ Å and $\varepsilon_M=5.5$ ($s=2.3v_F$), and $a=3.1$ Å and $\varepsilon_M=8$ ($s=2v_F$), respectively. For the values of the parameters for La-Sr-Cu-O, the linear dependence extends up to energies of order 0.5 eV.

may be approximated by a sum of two δ functions. This implies that one may replace, with reasonable accuracy, the plasmon band in the reciprocal q_z space by just two branches: the upper (U) branch, which is similar to the usual 3D dispersion, and a lower branch (L) which does not have a gap at $\kappa=0$.

The dependence $\Omega_{\rm pl}(\kappa,q_z)$ can be rewritten in the form (we focus on the L branch):

$$\Omega_{\rm pl}=v_F\kappa G(\kappa c)\ ,$$
$$G(x)=\left[1+\left[\frac{2F(\kappa,\pi)}{r_B\kappa}\right]^2\frac{1}{1+\dfrac{4F(\kappa,\pi)}{r_B\kappa}}\right]^{1/2};\quad F(\kappa,\pi)=\frac{\sinh(\kappa c)}{\cosh(\kappa c)+1}\ , \tag{19}$$
$$\lim_{\kappa\to 0}F(\kappa,\pi)=\kappa c\ .$$

We used the relation $p_F=(2cn)^{1/2}h$ (see, e.g., Ref. 9). In the region of small κ, $G(0)=(1+a)^{1/2}$; therefore $\Omega_{\rm pl}=v_F\kappa(1+a)^{1/2}, a=2c/r_B$.

Let us estimate the value of the slope (group velocity of the L branch) proportional to the first derivative of $\Omega_{\rm pl}$ for small κ. It is apparent from Eq. (19) that it is constant for small κ: $\Omega_{\rm pl}=s\kappa$; $s=(1+a)^{1/2}v_F$. For La-Sr-Cu-O we have $c=6.5$ A, $m^*\simeq 4m_e$ (see Ref. 9). There is some uncertainty about the value of ε_M, which has a value between 4.5 and 8 (Bozovic, unpublished). As a result the slope is equal to $(3-4)v_F$. The function $G(x)$, calculated for La-Sr-Cu-O, is presented in Fig. 1.

Note that the value of v_F in La-Sr-Cu-O is small ($v_F\simeq 8\times 10^6$ cm^{-1}); see (9). Therefore, the slope of the L branch is also relatively small compared to the Fermi velocity of conventional metals.

As a result, we arrive at the following expression for the energy loss per unit time γ:

$$\gamma\equiv\frac{\partial E}{\partial t}=\frac{1}{4\pi^3}\frac{1}{v_\parallel c}\sum_{i=L,U}\int dq_\parallel\Omega_i^2|V_{q_\parallel,q_z^i}|^2\tilde{N}_i\Pi\times\left[\frac{q_\parallel v_F}{\Omega_i}\right]\frac{1}{\sin\phi}\bigg|_{\Omega=\Omega_i}\ . \tag{20}$$

Here $q_z^L=\pi/c$, $q_z^U=0$ (see Refs. 5), $\tilde{N}_i=[1-\exp(-\beta\Omega_i)^{-1}]$, and $\sin\phi$ can be evaluated from Eqs. (17)–(19).

The function $\Omega_i(\kappa,q_z)$ is defined by Eqs. (17)–(19).

One can see directly from Eq. (20) that the total loss γ consists of two parts: $\gamma=\gamma_U+\gamma_L$. In this paper, we focus on the temperature dependence of γ. This dependence arises from the factor $\tilde{N}$, which is related to the thermal occupation N ($\tilde{N}=N+1$) of the L plasmon branch. For the U plasmon branch, only the zero-point motion contributes as the thermal occupation of this branch is very small (e.g., for the high-T_c oxides $\Omega_U\approx 1$ eV) and consequently γ_U does not depend on temperature. This is quite analogous to the ordinary case of temperature-independent EELS in the usual isotropic 3D case (cf. Refs. 1 and 6). The inequality $\omega_{\rm pl}\gg k_BT$ leads to temperature-independent EELS.

Now consider the term γ_L. The presence of the lower branch leads to an entirely different result. By a change of variables $q_\parallel$ to $\omega(q_\parallel)$, we obtain

$$\gamma_L=\frac{1}{4\pi^3}\frac{1}{v_\parallel c}\int d\omega_L(dq_\parallel/d\omega_L)\times V^2_{q_\parallel,\pi/c}\omega_L^2\Pi\frac{1}{\sin\phi}\bigg|_{\omega=\omega_L}\tilde{N}\ , \tag{21}$$

where $V_{q_\parallel,\pi/c}$ is defined by Eq. (10). The derivative $(dq_\parallel/d\omega_L)$ can be calculated from the dispersion relation [Eq. (17)] and it allows us to evaluate the integral (21). It can be seen directly from Eq. (21) that the presence of the factor $\tilde{N}$ leads to the temperature dependence of γ_L.

It is convenient to introduce the dimensionless variable $x=\beta\omega_L$ and we obtain

$$\gamma_L=AT^3f(T)\ , \tag{22}$$

where $A=(4\pi^{-3}v_Fv_\parallel c)^{-1}$, and

$$f(T)=\int dx\,x^2(1-e^{-x})^{-1}F(x,T)\ , \tag{23}$$

where $F=V^2_{q_\parallel,\pi/c}\Pi(\sin\phi)^{-1}$.

The factor F can be expressed in terms of x with the use of Eq. (17). One should also note that F contains the direct dependence on $v_\parallel$ and v_z, the components of the velocity of the charged particle parallel and perpendicular to the layers.

Consider the most important case, namely $q_\parallel\ll\pi/c$. This case corresponds to the plasmon frequency $\omega_L\ll\tilde{\omega},\tilde{\omega}\approx\omega_U\approx(4\pi ne^2/m^*)^{1/2}$. Then with good accuracy one may neglect the dependence of $V_{q_\parallel,\pi/c}$ on $q_\parallel$, and put $\omega_L\sim sq_\parallel, s\approx v_F$. Assuming also that $v_\parallel\gg v_F$ (then $\sin\phi\approx 1$) (cf. Refs. 1 and 6), we obtain that $f(T)\sim\omega$ and

$$\gamma_L\sim T^3\ . \tag{22'}$$

This dependence is valid for $T\ll\tilde{\omega}$. For example, for La-Sr-Cu-O this range extends to $T\approx 10^3$ K.

The subsequent increase in T leads to a deviation from the simple dependence $\gamma\sim T^3$; namely, in the region $T\sim\tilde{\omega}$, there is a maximum of $\gamma_L(T)$ due to the factor $q_\parallel^{-2}$ in $V_{q_\parallel,\pi/c}$. However, this situation corresponds to temperatures which seem experimentally inaccessible.

Hence, the total losses are described by the sum $\gamma=\gamma_U+\gamma_L(T)$, where $\gamma_L\approx R^*(v_F/v_\parallel)(v_F/c)(T/\tilde{\omega})^3$ or

$$\Gamma=\Gamma_U[1+\alpha(c/a)^4(T/\omega)^3]\ ,$$

where c is the interlayer distance and a the intralayer lattice constant, $\tilde{\omega} \simeq v_F \pi / c$, $\alpha \simeq 1$; in La-Sr-Cu-O $c \simeq 6.5$ A, $a \simeq 3.8$ Å, $v_F \simeq 8 \times 10^6$ cm sec^{-1}; γ_U is a typical value for temperature-independent EELS (see, e.g., Ref. 10) (cf. Ref. 1), $R^* = m^* e^4 / \varepsilon_M^2 \hbar^2$. The temperature independence of γ_U allows a clear separation from γ_L.

The appearance of a temperature-dependent term in the expression for the energy loss is a novel effect in EELS. This feature is a direct consequence of the structure in the layer plasmon bands typical of these layered materials.

Finally, we want to make two comments. First, the same treatment we have developed in this paper is valid for positron-energy-loss spectroscopy (PELS). In that case there is no need to take the exchange effect into account (for EELS this can be done by the same method as in Ref. 6).

Secondly, the passing particle can interact not only with the electronic subsystem of the material, but also with the phonon subsystem. In the temperature region $T < \Theta_D$, it will also lead to a temperature-dependent loss feature (for $T > \Theta_D$ the temperature dependence of the EELS is due to plasmons only). However, this effect is small. Indeed, the matrix elements describing the interaction of the fast passing particle with phonons contains a nuclear displacement; in other words, it is proportional to a small, nonadiabatic parameter $\kappa a / L \ll 1$ (see, e.g., Ref. 11), where a is the amplitude of the vibration and L the lattice parameter. Therefore, the major contribution to EELS is due to the particle-layer plasmon interaction.

The presence of overlapping energy bands with sufficiently different masses leads to the appearance of the additional plasmon branch, which also does not have an energy gap at $q \to 0$ (see, e.g., Refs. 12 and 13). This branch exists even in the usual 3D systems. This branch can be treated in exactly the same fashion as the L branch and would lead to a further temperature-dependent term γ_D in EELS. However, a large value of the group velocity leads to a relatively small value of γ_D. Nevertheless, the branch in the usual 3D systems with suitable overlapping bands may be detected by the use of temperature-dependent EELS (PLS). Another interesting class of materials with an "acoustic" plasmon branch[14,15] is found in the quasi-1D organic charge transfer conductors.[16] These questions will be discussed in detail elsewhere.

In this paper we have given a theoretical treatment of electron-loss spectroscopy in layered conductors. The main result of this study is the prediction of a novel temperature-dependent feature of EELS. This temperature dependence is caused by the unusual structure of the plasmon band. This structure, and, in particular, the existence of the low energy plasmon branch, can be identified by EELS. We suggest that experiments designed to search for temperature-dependent loss features by EELS or PELS would be very interesting and important for the confirmation of the results presented here.

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APPENDIX

In this appendix we derive the layer plasmon dispersion relations [Eq. (17)] and discuss their relationship to both the hydrodynamic [Ref. 5(b)] and many-body (RPA) derivations [Refs. 5(d)–(g)].

The collective electronic modes of the layered electron gas (LEG) are solutions (within RPA) of the set of equations

$$1 + v(\kappa, q_z)\pi^0(\kappa, \omega) \equiv 0 , \tag{A1}$$

where the interaction

$$v(\kappa, q_z) = 2\pi e^2 / \kappa \varepsilon_M F(\kappa, q_z) ;$$

$$F(\kappa, q_z) = \frac{\sinh(\kappa c)}{\cosh(\kappa c) - \cos(q_z c)}$$

arises from the layered structure and $\pi^0(\kappa, \omega)$ is the polarizability of a single layer[4,8] given by

$$\pi^0(\kappa, \omega) = \frac{m^*}{\pi} \left\{ \left[1 - \left[\frac{v_F \kappa}{\omega} \right]^2 \right]^{-1/2} - 1 \right\} . \tag{A2}$$

Equation (A1) can be solved algebraically and the explicit solutions have the form

$$\Omega_p(\kappa, q_z) = v_F \kappa \left[1 + \left[\frac{2F}{r_B \kappa} \right]^2 \frac{1}{1 + \frac{4F}{r_B \kappa}} \right]^{1/2} . \tag{A3}$$

Here the function F [the layer form factor in the terminology of Ref. 5(b)] appears in both the numerator and denominator of the second term [r_B is the effective Bohr radius for the carrier defined in Eq. (18) of the text]. We note that in the hydrodynamic approximation, the correction to the compressibility term proportional to $(v_F \kappa)^2$ appears only as an additive term. It is a consequence of the RPA approximation, which sums a particular subset of polarization diagrams and gives the (to our knowledge) new result [Eq. (A3)].

We note that it is common in the literature to expand Eq. (A2), making the high frequency approximation $\omega \gg v_F \kappa$ and find

$$\Omega_P = \left[\frac{2 v_F^2 \kappa F}{r_B} \right]^{1/2} . \tag{A4}$$

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