New mechanism for spin relaxation during light scattering

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A theory is derived for the universal relaxation lineshape in quasielastic electron scattering of light by spin-density fluctuations in the multicomponent spin systems in intermetallic actinides with heavy fermions, high-T_c superconducting crystals, and heavily doped semiconductors. A new spin relaxation mechanism is proposed. This mechanism provides a physical interpretation of the width of the quasielastic-scattering line, which is independent of the light wave vector. It also explains the high intensity of this line. Possibilities for estimating certain microstructural parameters of intermetallic actinides—the energies of the spin–orbit splitting and of optical phonons—from spectra are discussed.

1. INTRODUCTION

In connection with the search for high-T_c superconductors, much experimental information has recently been accumulated on light scattering by free electrons in the intermetallic compounds UPt_3 (Ref. 1), UBe_{13} (Refs. 2 and 3), and URu_2Si_2 (Ref. 4). These intermetallic actinides, all of which have heavy fermions, differ in terms of many microscopic parameters (structure, composition, degree of localization of the $5f$ electrons, and the presence of a magnetic order). Their Fermi surfaces are completely different. Nevertheless, a scattering of light by spin-density fluctuations has been observed in all the actinides which have been studied. This is true despite the fact that magnetic scattering is known to be weak[5] and despite the difficulties in detecting scattering in nontransparent media.

Much is unclear about the behavior of heavy fermions.[6] For example, there is a contradiction between their finite contribution to the conductivity, even the presence of a superconducting pairing, and the local nature of their magnetic moments. Electron scattering of light, which has previously eluded observation in normal metals, according to Ref. 7, has unexpectedly turned out to be a convenient tool for studying intermetallic actinides. According to Ref. 8, for example, the observation of a scalar scattering by fluctuations of the electron density in UPt_3 (Ref. 9), combined with the absence of the scattering from UBe_{13} (Ref. 8), suggests that the degree of localization of the $5f$ electrons is greater in the second of these compounds.[8] In contrast with scalar scattering, skew scattering by spin-density fluctuations has been detected in all the actinides which have been studied.[8] A Lorentzian lineshape has been observed everywhere, with a linewidth Γ_S which is independent of the wave vector of the light. This result is evidence of an intense spin relaxation. It was concluded on this basis in Ref. 8 that the magnetic moments of the $5f$ electrons are completely localized, but the nature of this localization remains unclear. It is believed that the spin relaxation frequency is governed primarily by collective effects such as the Korringa and Kondo effects (Ref. 10, for example). The Korringa coefficient a found in Ref. 3 from the slope of the linear part of the temperature dependence $\Gamma_S(T)$ turned out to be an order of magnitude higher than the theoretical predictions.

In this letter we wish to propose a special spin relaxation mechanism which is unrelated to collective effects and which requires only the presence of ordinary impurities in the crystal. A necessary condition for the manifestation of this mechanism in heavily doped semiconductors is that several (at least two) nonequivalent valleys be filled with electrons. An electron distribution of this sort has been achieved in Ge through heavy doping,[11] and in GaAs by the application of hydrostatic pressure.[12] The intermetallic actinides which have been studied to date, as well as the high-T_c superconductors, have several partially filled electron subbands: bands of heavy and light fermions in actinides[6] and conducting chains and planes in the high-T_c superconductors. These compounds are crystals of modest quality or polycrystalline materials. Accordingly, the spin relaxation mechanism which we are proposing here should be considered in the case of actinides also.

2. RELAXATION THEORY OF THE LINESHAPE IN THE SCATTERING SPECTRUM

To calculate the cross section for scattering by spin-density fluctuations, we need to find the antisymmetric part of the electric susceptibility tensor.[5,13] If, for definiteness, we conduct the discussion in terms of intermetallic actinides, this antisymmetric part of the tensor should be linked with fluctuations of the axial magnetization vector.[1,5] In this case the scattering cross section can be written[1]

$$\frac{\partial^2\sigma}{\partial\omega_S\partial\Omega}=Vr_0^2[\mathbf{e}_I\cdot\mathbf{e}_S]^2\int_{-\infty}^{\infty}dte^{i(\omega_I-\omega_S)t}\langle M_z(t)M_z(0)\rangle. \tag{1}$$

Here $\langle...\rangle$ mean a statistical average over initial states, $r_0=e^2/mc^2$ is the classical radius of the electron, Ω is the solid angle, V is the volume of the scattering part of the crystal, $\mathbf{e}_I$ and ω_I are the polarization vector and frequency of the incident light, $\mathbf{e}_S$ and ω_S are the corresponding properties of the scattered light, and M is the dimensionless magnetization, whose projection M_z is[10]

$$M_z=\sum_\alpha R_\alpha(\delta n_\uparrow^{(\alpha)}-\delta_\downarrow^{(\alpha)}). \tag{2}$$

Here $\delta n_{\uparrow(\downarrow)}^{(\alpha)}$ is a fluctuation of the concentration of the fermions of species α with spin up (down), and R_α is a resonant factor, which depends on the electronic band structure. Through a collision with a phonon or impurity, a heavy fermion can go into a different band over a time scale τ, preserving its spin and becoming an ordinary conduction electron. The equalization of the concentrations of the different fermions which is caused by processes of this type is described by ordinary relaxation equations. The corresponding relaxation lineshape is known well for the case of scattering by fluctuations of the spin density in multivalley semiconductors.[14] Ignoring fermion-diffusion processes, in accordance with experiments,[1–4,8,9] we write the following relaxation equation for the relative population of the spin states:

$$\frac{\partial(\delta n_\uparrow^{(\alpha)}-\delta n_\downarrow^{(\alpha)})}{\partial t}=\frac{1}{\tau}\sum_\beta I_{\alpha\beta}(\delta n_\uparrow^{(\beta)}-\delta n_\downarrow^{(\beta)}). \tag{3}$$

The symmetric collision matrix $I_{\alpha\beta}$ reduces here to the difference between "outgoing" and "incoming" terms, which are proportional to the densities of the final states according to Ref. 15:

$$I_{\alpha\beta}=\left(\frac{\partial n_\alpha}{\partial\zeta}\right)_T\left[\delta_{\alpha\beta}-\frac{(\partial n_\beta/\partial\zeta)_T}{(\partial n/\partial\zeta)_T}\right], \tag{4}$$

where n_α is the equilibrium concentration of the fermions of species α,

$$n=\sum_\alpha n_\alpha \tag{5}$$

is their total concentration, and ζ is the chemical potential. Simple relaxation equations (3) and (4) hold regardless of the degree to which the particles of one species or another are localized. In the model of alternating superconducting and nonsuperconducting layers proposed by Abrikosov for high-T_c superconductors,[16] system of equations (3) is an infinite number of equations, which are coupled through $I_{\alpha\beta}$. In this case, the role of τ is played by the time scale of the transition between neighboring conducting layers, e.g., between CuO chains and CuO_2 planes. It was shown in Ref. 17 that the spectral correlation function

$$\psi_\alpha(\omega)=\left(\frac{\partial\zeta}{\partial n_\alpha}\right)_T\sum_\beta R_\beta\int_0^\infty dt e^{i(\omega_I-\omega_S)t}\langle[\delta n_\uparrow^{(\alpha)}(t)-\delta n_\downarrow^{(\alpha)}(t)]$$
$$\times[\delta n_\uparrow^{(\beta)}(0)-\delta n_\downarrow^{(\beta)}(0)]\rangle, \tag{6}$$

which determines the mean square fluctuation of the magnetization and also the scattering cross section, according to Eqs. (1) and (2), satisfies the same equations as are satisfied by the fluctuating quantity $\delta n_\uparrow^{(\alpha)}-\delta n_\downarrow^{(\alpha)}$ itself. A system of equations for the functions in (6) can be found from (3) and (4) by taking one-sided Fourier time transforms and incorporating the initial conditions (Ref. 15, for example):

$$\langle(\delta n_\uparrow^{(\alpha)}-\delta n_\downarrow^{(\alpha)})(\delta n_\uparrow^{(\beta)}-\delta n_\downarrow^{(\beta)})\rangle=T\left(\frac{\partial n_\alpha}{\partial\zeta}\right)_T\delta_{\alpha\beta}. \tag{7}$$

In the simple case in which, in addition to the heavy fermions ($\alpha=h$) , we consider electrons of only one band, which play the role of the light particles ($\alpha=l$), the system of equations for the functions in (6) becomes

$$-i\omega\psi_l+\frac{1}{\tau}\frac{(\partial n_h/\partial\zeta)_T}{(\partial n/\partial\zeta)_T}(\psi_l-\psi_h)=TR_l, \tag{8}$$

$$-i\omega\psi_h+\frac{1}{\tau}\frac{(\partial n_l/\partial\zeta)_T}{(\partial n/\partial\zeta)_T}(\psi_h-\psi_l)=TR_h, \tag{9}$$

where $\omega=\omega_I-\omega_S$. Substituting the solution of this system (ψ_l, ψ_h) into (1) and (2), and using definition (6), we find the following expression for the cross section:

$$\frac{\partial^2\sigma}{\partial\omega_S\partial\Omega}=TVr_0^2\frac{(\partial n_l/\partial\zeta)_T(\partial n_h/\partial\zeta)_T}{(\partial n/\partial\zeta)_T}[\mathbf{e}_\uparrow\cdot\mathbf{e}_S]^2\frac{\tau}{1+(\omega\tau)^2}(R_h-R_l)^2. \tag{10}$$

It follows from (10) that the cross section has a Lorentzian relaxation spectral shape, which is the same for all intermetallic actinides, high-T_c superconducting crystals, and multivalley semiconductors. The constant value $\Gamma_S=1/\tau$, which agrees with experiment, is a consequence of relaxation equations (8) and (9), which hold regardless of the degree to which the magnetic moments of the heavy fermions are localized. Consequently, there is no basis for the assertion in Ref. 8 that the detection of a constant value of Γ_S, which does not depend on the wave vector of the light, is evidence of a complete localization of the magnetic moments of the heavy fermions. The numerous examples of observations of a relaxation lineshape with a constant width $\Gamma_S=1/\tau$ in ideal solid-state plasma and Fermi liquids[15] are evidence for the opposite conclusion.

3. ABSOLUTE SCATTERING INTENSITY

It follows from (10) that observing the relaxation contribution to the scattering by spin-density fluctuations requires a significant density of states in both of the subbands involved. In addition, the absolute scattering intensity is determined by the difference between the resonant-gain coefficients for the different subbands: $\Delta R=R_h-R_l$. At present, explicit expressions for these coefficients are available only in the case of isolated atoms[10] and semiconductors.[12] For n-type semiconductors (Ge, GaAs, and InSb), the coefficients R_α are[12]

$$R_\alpha=\hbar\omega_I\frac{2(P_{cv}^{(\alpha)})^2}{3m}\frac{\Delta_\alpha(\Delta_\alpha+2E_{g\alpha})}{[E_{g\alpha}^2-(\hbar\omega_I)^2][(E_{g\alpha}+\Delta_\alpha)^2-(\hbar\omega_I)^2]}, \tag{11}$$

where $E_{g\alpha}$ and $P_{cv}^{(\alpha)}$ are parameters of the band structure of the semiconductor (Ref. 18, for example), and Δ_α is the energy of the spin–orbit splitting of the valence band of the semiconductor in the corresponding region of the Brillouin zone. It can be seen from (11) that the difference between the resonant-gain coefficients $R_l\neq R_h$ for the different subbands arises primarily because of the difference between the energies for spin–orbit splitting, $\Delta_l\neq\Delta_h$. In the semiconductors n-Ge and n-GaAs, with a relatively weak spin–orbit coupling for all valleys, we have $\Delta_\alpha\ll E_g$ and thus

$$\Delta R\approx\frac{(\Delta_\Gamma-\Delta_L)\hbar\omega_I}{E_g^2-(\hbar\omega_I)^2}\ll R_\alpha. \tag{12}$$

In InSb, however, the opposite inequality, $\Delta_\Gamma\gg E_g$, holds for the central Γ valley, so we have $\Delta R\approx R_\alpha$. Consequently, the resonant amplification makes it possible to achieve the inequality $\Delta R\gg 1$, under which the process with cross section (10) outweighs all other processes. The electronic band structure of intermetallic actinides has not been studied adequately; we do not know concrete values of the coefficients in (11) for these actinides.

Because of the high atomic numbers of the constituent elements, one might suggest that there is a strong spin–orbit coupling, which leads to satisfaction of the condition $\Delta R \gg 1$. From the intensity of the quasielastic skew scattering one can draw conclusions about the relative role played by a spin–orbit splitting of electron bands. It follows from the discussion above that this role is significant in intermetallic compounds. In crystals of high-T_c superconductors, in contrast, a scattering of this type has yet to be observed, so one might suggest that the spin–orbit splitting plays a minor role here.

From the width of the quasielastic-scattering spectrum, (10), one can determine the temperature dependence of various relaxation processes. Working from the beginning of the linear region on the temperature dependence $\Gamma_S(T)$, we can estimate the frequency of the optical phonon responsible for the relaxation time τ on the basis of Refs. 3 and 8: $\omega_{\rm opt} \approx 5$ meV. The phonon spectrum of intermetallic actinides is also unknown. Since the frameworks of their lattices consist of nuclei of fairly heavy elements, and since there are many atoms in a unit cell, one might expect the appearance of some fairly soft phonons, which would correspond to the estimate given above.

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